STACKS

# JOURNAL of the AIR POLLUTION CONTROL ASSOCIATION

*Founded 1907*

Dr. Paul Clifford Cross, President, Mellon Institute (see page 441).

**September 1961**
**Vol. 11, No. 9**

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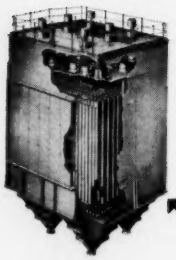

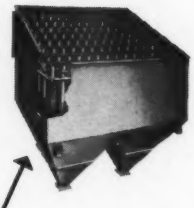

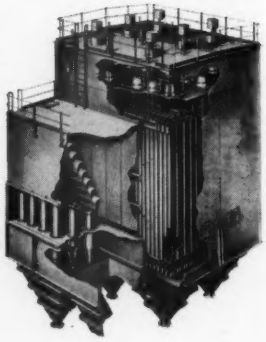

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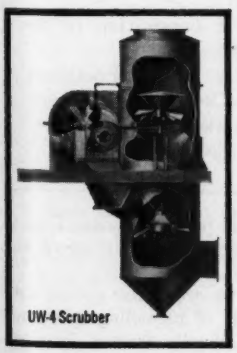

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## LETTERS to the EDITOR

### John Wood Brochure

Mr. Loree Collins
John Wood Company
Florham Park, New Jersey

Dear Mr. Collins:

This will acknowledge receipt of your letter of April 12, 1961, transmitting a copy of the brochure entitled "Together We Can Check the Blight of Air Pollution."

Because of our deep interest in the problems of air pollution, we have read your brochure with considerable care. I am sure that we are in general agreement as to the objectives which are outlined concerning the need for more effective air pollution programs and the thought that much more can and must be accomplished in relation to air pollution control.

With respect to the means for accomplishing these objectives, however, there are, in our opinion, serious questions which can be raised, and the report has some inaccuracies and omissions which may tend to mislead the reader. Without attempting to cover all of the details of this kind, we would like to comment on several such items as follows:

*A.* In the President's message on natural resources, outlining the need for greater Federal activity in relation to air pollution, the President also stated that this "new leadership, research, and financial and technical assistance for the control of air pollution" would be located in a "special unit within the Public Health Service, under the Department of Health, Education, and Welfare."

*B.* The suggestion for a policy that Federal installations across the Nation should observe good practices in relation to air pollution control has, of course, already been adopted and is included in Public Law 84-139, as amended. If there are widespread deviations from this policy, we would appreciate being informed of them, in order that appropriate action might be initiated.

*C.* The suggestion for segmentation of the Federal air pollution program activities is open to serious question, in our opinion. At present, although several agencies such as the Atomic Energy Commission and the Department of Defense carry on air pollution activities in connection with the operation of their facilities, the elements of the over-all Federal research, technical assistance, and training activities in relation to air pollution are unified within the Public Health Service. As part of this comprehensive program, the service of several other Federal agencies, e.g., the Weather Bureau, Bureau of Mines, and National Bureau of Standards, are utilized and are not separate activities, as one might infer from the brochure. We believe, in general, that splitting of the existing unified Federal program would tend to weaken it rather than providing strength.

*D.* While we are in full accord with the desirability of providing additional personnel specifically trained for dealing with air pollution problems, we have serious reservation as to the mechanism proposed for supporting such training and of the degree of regimentation of the university edu-

(*Continued on p. 402*)

**Secretary's Page**

## No Summer Let-Up

After each year's annual meeting, there is usually a lull in the correspondence load at headquarters. Vacation time for most of our members plus the general let-down after the annual meeting, allows personnel in Pittsburgh to catch up on filing, routine matters, and a little breather before the merry-go-round starts again. Not this year. So much interest in APCA, its annual meeting papers, its publications, membership, and technical information requests that the work-load has doubled in number of letters received and answered over the equivalent period last year. In addition, members seem to be quite pleased that they have received pre-prints of annual meeting papers and complete sets of the 74 out of 78 pre-prints available within 30 days of the annual meeting. The decision to do away with proceedings that are not published until nine months after the annual meeting, but rather make available as many pre-prints as possible in unbound sets in this length of time has been an extremely popular one. Your executive secretary has had inquiries from technical organizations 10 times our size and larger about the techniques for handling this type of pre-print operation. The answer is—prior planning.

### Board Actions

Some of the important actions of the Board of Directors during the June 11 and June 12 meetings of the Board were:

*1* Heard a report on increased activity concerning the International Affairs committee with emphasis on the projected transatlantic telephone conversation with the members of the National Society for Clean Air of Great Britain scheduled for October 5, 1961. (See story in another part of JOURNAL.)

*2* Approved publication of the Governmental Agencies Directory and APCA Membership Directory for 1961–62.

*3* Extension of the APCA Abstracts contract with the U. S. Public Health Service was authorized for the year 1961–62.

*4* Approved initiation of new by-law amendments authorizing formation of local sections and laying the ground rules for them.

*5* Accepted a report from incoming President Faith on Membership activities. As of June 30, the total membership has climbed to 2062—this does not include the 100 new members which will shortly be added with the MAPPA section affiliation.

*6* Accepted with modifications the Interim Report of the Future of APCA committee from Chairman R. L. Ireland.

*7* Authorized a new Technical Co-ordinating committee to be known as Ambient Air Standards Committee TR-5.

*8* Adopted a resolution in behalf of APCA urging President Kennedy, Secretary of HEW, and the Surgeon General to convene another National Conference on Air Pollution in 1962.

*9* Passed a resolution memorializing the late Ellis E. Smauder.

FOUNDED 1907

# JOURNAL OF THE AIR POLLUTION CONTROL ASSOCIATION

A Monthly Journal
Devoted to
Air Purification

Editor: Arnold Arch
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Assistant Editor: Pat Martin

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The Editor welcomes writers interested in air pollution problems and encourages them to use the pages of this journal for the expression of their opinions. But neither he nor the Association assumes responsibility for the validity of the opinions expressed herein.

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## LETTERS TO THE EDITOR

(*Continued from p. 400*)

cational process outlined in the report.

*E.* The tax incentive to industry which is proposed raises several fundamental problems. The desirability of using such incentives to stimulate the attainment of objectives considered socially desirable constitutes a basic question of Federal tax policy. Also, difficulty has been experienced in this Department in obtaining a reasonable basis for estimating the degree to which any such tax incentive would actually stimulate industry to further air pollution control installations; if you have data bearing on this subject, we would appreciate it greatly.

*F.* As a final comment, we question seriously the soundness of the proposal to assign responsibility and provide financial support to a non-Government organization for co-ordination of pollution abatement activities on the national level. In general, we would not consider this concept sound and believe that, if done, it would appear to be an abnegation of direct Federal responsibility.

In summary, we are in full agreement with the need for strengthening air pollution control activities throughout the Nation and the need for participation by industry, all levels of government, and other organizations concerned in this over-all effort. In furthering this objective, we believe that the general proposal and organization as outlined by the President is best suited to this purpose.

WILBUR J. COHEN
*Assistant Secretary*

Mr. Wilbur J. Cohen
Assistant Secretary
Department of Health, Education, and Welfare
330 Independence Avenue, S. W.
Washington, D. C.

Dear Mr. Secretary:

Thank you for your letter and comments regarding our brochure "Together We Can Check the Blight of Air Pollution."

We certainly didn't mean to mislead and perhaps our inaccuracies are more a matter of interpretation than misquoted facts.

In regard to your request for information with reference to tax incentives. I have written several interested parties soliciting data and comments. I shall, of course, send them along to you when I have collected them.

Thank you again for your comments and interest and if we can be of any assistance to you please don't hesitate to call on us.

LOREE COLLINS
*Vice-President*

(*Continued on p. 439*)

# RELATION of Benzene Soluble ORGANIC MATTER to Suspended PARTICULATE MATTER in the Atmosphere*

**ELBERT C. TABOR** and **DONALD H. FAIR**, National Air Sampling Network, Air Pollution Engineering Research, Robert A. Taft Sanitary Engineering Center, Cincinnati, Ohio

This paper reports the relationship between the organic fraction and the total weight of suspended particulate air pollutants based on data obtained in 1957–58 by the National Air Sampling Network from a large number of American cities and nonurban areas.[1] Network samples, collected biweekly on a random sampling schedule, represent the particulate pollutants from approximately 75,000 cubic feet of air. The air is filtered through 8 × 10 in. glass fiber filters, at an average of about 50 cfm for the 24-hour sampling period. In a previous paper[2] the use of the statistical parameters, geometric mean, and standard geometric deviation, for the delineation of various air pollutant levels was demonstrated.

In order to produce valid statistical parameters it is necessary to impose rather rigid restrictions on the data used. The criteria for inclusion in this study were the same as those used previously,[2] namely; in any one year at least 20 samples must have been collected at a given site in a city; these samples must be evenly distributed over a calendar year with all quarters represented by samples collected in two months of the quarter; with no more than three samples from any month and no more than seven from any quarter. For certain parts of this paper, in addition to meeting the criteria given above, the data have to further qualify by showing log-normal distribution.

In this paper the discussion will be limited to a consideration of the quantitative relations existing between levels of the benzene soluble portion of suspended air particulate matter and levels of suspended air particulate matter itself. For this discussion suspended air particulate matter can be defined as those components of the atmosphere which are collected on an 8 × 10 in. glass fiber filter when air is drawn through it at a rate of approximately 30–60 cfm and benzene soluble organic matter can be described as that organic material which is removed from suspended air particulate matter by extraction with boiling benzene, for six to eight hours, in a standard Soxhlet extraction apparatus.

Gross data for the period 1957–58 available for this study included data from 44 cities for 1957, from 48 cities for 1958, and from 64 additional cities for both years. After elimination of data which did not meet the previously listed criteria (not including the log-normality test), there remained for use in the paper, data from 42 cities for 1957, from 48 cities for 1958, and from 54 additional cities for both years, for a total of 144 cities. Satisfactory nonurban data were available from one station for 1957, from 34 stations for 1958, and from three stations for both years, for a total of 38 stations.

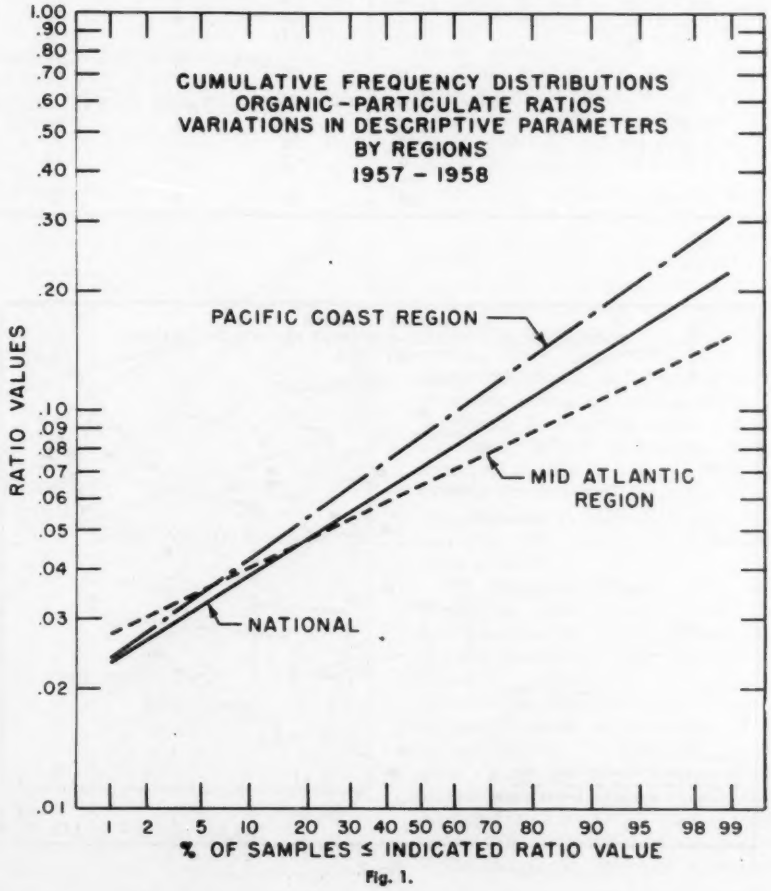


Fig. 1.

* Presented at the 53rd Annual Meeting of APCA, Netherland-Hilton Hotel, May 22–26, 1960, Cincinnati, Ohio.

**Table I—Comparison of Regional and National Data for Benzene Soluble Organic to Suspended Particulate Ratios**

| | No. of Cities | AM | GM | SGD | Middle 80% of Values 10%–90% | 90%/10% |
|---|---|---|---|---|---|---|
| National | 144 | 0.082 | 0.074 | 1.62 | 0.038–0.140 | 3.7 |
| New England | 18 | 0.090 | 0.083 | 1.47 | 0.049–0.137 | 2.8 |
| Mid Atlantic | 20 | 0.070 | 0.066 | 1.43 | 0.040–0.104 | 2.6 |
| Mid East | 13 | 0.089 | 0.080 | 1.51 | 0.042–0.141 | 3.4 |
| South East | 16 | 0.097 | 0.089 | 1.53 | 0.051–0.153 | 3.0 |
| Mid West | 27 | 0.075 | 0.069 | 1.54 | 0.042–0.141 | 3.4 |
| Great Plains | 12 | 0.063 | 0.056 | 1.67 | 0.029–0.102 | 3.5 |
| Gulf South | 13 | 0.075 | 0.066 | 1.75 | 0.032–0.130 | 4.1 |
| Rocky Mountain | 5 | 0.079 | 0.070 | 1.74 | 0.033–0.142 | 4.3 |
| Pacific Coast | 20 | 0.099 | 0.088 | 1.71 | 0.043–0.175 | 4.1 |

**Table II—Benzene Soluble to Suspended Particulate Ratios**

Classification of All Stations for 1957–58
by Geometric Mean and Standard Geometric Deviation

| Standard Geometric Deviation | Geometric Mean 0.028–0.049 | 0.050–0.073 | 0.074–0.111 | 0.112–0.140 | Total |
|---|---|---|---|---|---|
| 1957 (Biennial and triennial stations) | | | | | |
| 1.22–1.41 | 2 | 12 | 3 | — | 17 |
| 1.42–1.61 | 1 | 10 | 8 | — | 19 |
| 1.62–2.61 | 1 | 1 | — | 1 | 3 |
| Total | 4 | 23 | 11 | 1 | 39 |
| 1958 (Biennial and triennial stations) | | | | | |
| 1.22–1.41 | 1 | 4 | 15 | 3 | 23 |
| 1.42–1.61 | — | 2 | 13 | 4 | 19 |
| 1.62–2.61 | — | 2 | 1 | — | 3 |
| Total | 1 | 8 | 29 | 7 | 45 |
| 1957–58 (Every year stations) | | | | | |
| 1.22–1.41 | 1 | 4 | 8 | 1 | 14 |
| 1.42–1.61 | — | 11 | 16 | — | 27 |
| 1.62–2.61 | 4 | 6 | 2 | — | 12 |
| Total | 5 | 21 | 26 | 1 | 53 |

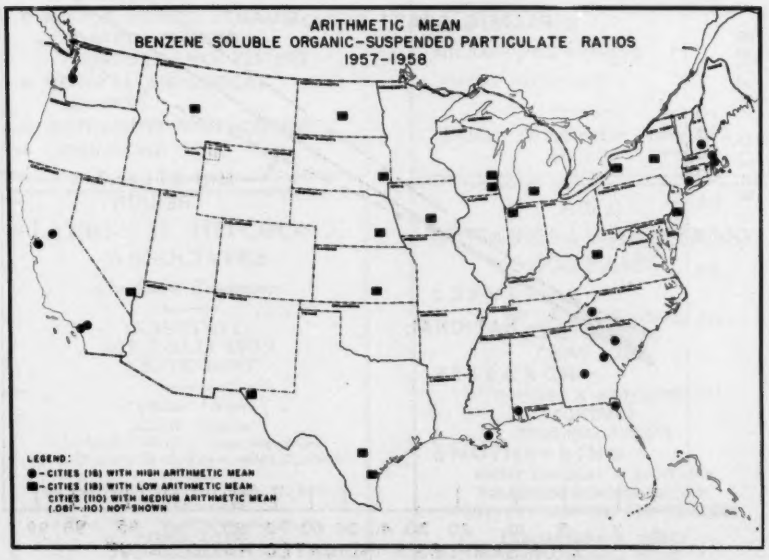
ARITHMETIC MEAN
BENZENE SOLUBLE ORGANIC–SUSPENDED PARTICULATE RATIOS
1957–1958
LEGEND:
● – CITIES (18) WITH HIGH ARITHMETIC MEAN
■ – CITIES (18) WITH LOW ARITHMETIC MEAN
CITIES (110) WITH MEDIUM ARITHMETIC MEAN
(.081–.110) NOT SHOWN

Fig. 2.

## Urban Relationships

Of the benzene soluble organic to suspended particulate ratio values for the 144 cities, 137 ratios passed the test for log-normality of distribution, leaving data from only seven (Brockton, El Paso, Evansville, Las Vegas, Peoria, St. Paul, and San Juan) not normally distributed. The ratios for the individual states, except Puerto Rico and Nevada, regions, and the nation were log-normally distributed. Of the every year stations, when both year's ratios are combined, only Las Vegas fell into the nonnormal category (Las Vegas fell into this category in each of these years also). Eight cities (Jackson, Miss., Detroit, Minneapolis, Houston, Dallas, Helena, Anchorage, and Los Angeles), in spite of falling into the nonnormal group for either one or both of the two years, were categorized as normally distributed on the basis of the composite two-year data. This emphasizes the need for collecting data for several years before attempting an exacting statistical analysis of such data.

In the case of the benzene soluble-suspended particulate ratios, the national arithmetic mean (AM) ratio was 0.082 or, in other words, the national average suspended particulate sample consisted of 8.2% benzene soluble materials (Table I). The statistical parameters determined for the nation as a whole, e.g., the geometric mean (GM) of 0.074, the standard geometric deviation (SGD) of 1.62, can be shown graphically (Fig. 1). The graph shows that the middle 80% of the ratio values fall between 0.038 and 0.140. By considering the middle 80% of the data, the influence of extremely high and/or low values is eliminated. The arithmetic mean ratios for the New England, Mid East, South East, and Pacific Coast regions were higher, and those of the Mid Atlantic, Mid West, Great Plains, Gulf South, and Rocky Mountain regions were lower than the national. When the descriptive parameters (GM and SGD) are examined on a regional basis, some differences can be observed. In the Pacific Coast region, with a high GM and high SGD, we find the most extreme range of values. The Great Plains, Gulf South, and Rocky Mountain follow the same general pattern, except for slightly lower GM values. The Mid Atlantic region, with the smallest SGD and fairly low GM, shows the narrowest range of values. The remaining regions fall somewhere between these extremes.

For the 144 individual cities we find a range of 0.036–0.151 for the arithmetic mean ratios. Seventy-one cities have mean ratios equal to, or above, the national mean of 0.082, and 73 are below, indicating a normal distribution of values. Locations of cities with

RATIO VALUES

**Table III—Benzene Soluble to Suspended Particulate Ratios**

Classification of Every Year Stations for 1957–58 by Geometric Mean and Standard Geometric Deviation

| Standard Geometric Deviation | Geometric Mean 0.028–0.049 | 0.050–0.073 | 0.074–0.111 | 0.112–0.140 | Total |
|---|---|---|---|---|---|
| | | 1957 only | | | |
| 1.22–1.41 | 1 | 10 | 2 | — | 13 |
| 1.42–1.61 | 6 | 9 | 8 | — | 23 |
| 1.62–2.11 | 2 | 5 | 2 | — | 9 |
| Total | 9 | 24 | 12 | | 45 |
| | | 1958 only | | | |
| 1.22–1.41 | — | 1 | 20 | 4 | 25 |
| 1.42–1.61 | — | 6 | 8 | 3 | 17 |
| 1.62–2.11 | — | 2 | 1 | — | 3 |
| Total | — | 9 | 29 | 7 | 45 |
| | | 1957–58 | | | |
| 1.22–1.41 | — | 3 | 6 | — | 9 |
| 1.42–1.61 | 1 | 11 | 18 | — | 30 |
| 1.62–2.11 | 3 | 1 | 2 | — | 6 |
| Total | 4 | 15 | 26 | — | 45 |

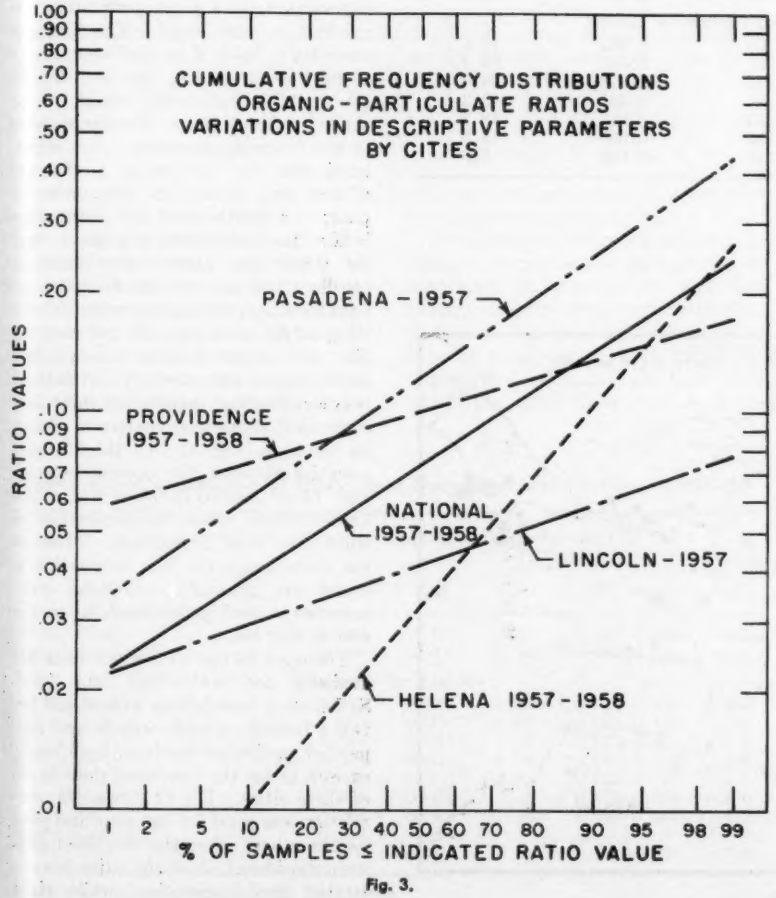


Fig. 3.

high mean ratios and those with low ratios are shown in Fig. 2. Cities with high mean ratios are located mostly on or near the east, west, and Gulf coasts, in contrast to inland locations for cities with low mean ratios.

Communities having log-normally distributed ratio values are classified by the descriptive parameters, geometric mean, and standard geometric deviation, by individual years and for both years (Tables II and III). Table II includes those cities (137) showing log-normal distribution of data for either year, or 1957–58 combined. Table III includes only those cities (45) for which data for both single years as well as the combined years are log-normally distributed. One striking fact stands out in both tables; 80% of the single year cities and 84% of the two-year cities have ratios between a geometric mean of 0.05 and 0.11, and between standard geometric deviations of 1.22 and 1.61. There was a pronounced shift in 1958 towards higher geometric mean values. At first it might be concluded that this shift was due to a difference in the cities included in 1958 as compared to 1957, but on inspection of the table for the every year cities, the same shift is evident.

Extreme deviations by individual cities from the national norm are shown in Table IV, and presented graphically in Fig. 3. To illustrate these four extreme deviations, data were selected to show (*1*) high GM and high SGD (Pasadena), (*2*) high GM and low SGD (Providence), (*3*) low GM and high SGD (Helena), and (*4*) low GM and low SGD (Lincoln).

For those cities having log-normal distributions of benzene soluble organics to suspended particulate ratios, it is possible to predict occasional extreme values by the use of the geometric means and standard geometric deviations of both the ratios and the particulate levels. Those communities with high geometric means and high standard geometric deviations, could, on occasion, experience a situation where a fairly large part of the particulate matter would be organic in nature. That the extreme city in the nation in this regard was Pasadena is significant, since this is a city with a severe photochemical smog problem.

The foregoing discussion has considered the ratios existing between the concentrations of two pollutants and the distribution of these ratios. No indication is given, however, as to the constancy of relations between the two parameters under consideration. This is expressed by the correlation coefficient, e.g., if, on determination of the percent organic matter in the particulate samples from a station, it is found to be always 10%, regardless of the weight of the sample, it would be concluded

**Table IV—Comparison of Data from Cities Showing Greatest Deviation from National Norm**

| | Benzene Soluble Organic to Suspended Particulate Ratios | | | |
|---|---|---|---|---|
| | GM | SGD | —Middle 80% of Values— 10%–90% | 90%/10% |
| National | 0.074 | 1.62 | 0.038–0.140 | 3.7 |
| Pasadena, Calif. | 0.114 | 1.64 | 0.063–0.25 | 4 |
| Helena, Mont. | 0.050 | 2.61 | 0.010–0.110 | 11.0 |
| Providence, R. I. | 0.099 | 1.41 | 0.076–0.135 | 1.8 |
| Lincoln, Nebr. | 0.043 | 1.27 | 0.029–0.058 | 2.0 |

**Table V—95% Confidence Limits of Correlation Coefficients**

| Sample Size = 25 | | |
|---|---|---|
| Correlation Coefficient | Lower Limit | Upper Limit |
| 0.0 | −0.41 | +0.39 |
| 0.1 | −0.30 | +0.46 |
| 0.2 | −0.20 | +0.55 |
| 0.3 | −0.10 | +0.61 |
| 0.4 | 0.0 | +0.67 |
| 0.5 | +0.12 | +0.74 |
| 0.6 | +0.25 | +0.80 |
| 0.7 | +0.40 | +0.85 |
| 0.8 | +0.57 | +0.90 |
| 0.9 | +0.75 | +0.95 |

**Table VI—Comparison of Cities with High Values of Organic to Particulate Ratios with Those Having High Particulate Levels**

| | | I | II | I × II |
|---|---|---|---|---|
| City | Year | Mean Suspended Particulate Level μg/m³ | Organic to Particulate Ratio | Organic Level μg/m³ |
| Cities with High Organic to Particulate Ratios | | | | |
| Brockton | 1958 | 92 | 0.151 | 13.9 |
| Oakland | 1958 | 104 | 0.145 | 15.1 |
| Tacoma | 1958 | 111 | 0.141 | 15.6 |
| Los Angeles | 1958 | 216 | 0.140 | 30.2 |
| Sacramento | 1958 | 112 | 0.136 | 15.2 |
| Honolulu | 1958 | 52 | 0.131 | 6.8 |
| Mobile | 1958 | 184 | 0.130 | 24.0 |
| New Orleans | 1958 | 100 | 0.129 | 12.9 |
| Cities with High Suspended Particulate Levels | | | | |
| Gary | 1958 | 306 | 0.062 | 19.0 |
| East St. Louis | 1958 | 251 | 0.097 | 24.3 |
| Chattanooga | 1958 | 245 | 0.098 | 24.0 |
| Buffalo | 1957 | 241 | 0.051 | 12.3 |
| San Bernardino | 1957 | 235 | 0.082 | 19.2 |
| Phoenix | 1957–58 | 225 | 0.069 | 15.5 |
| El Paso | 1957 | 223 | 0.036 | 8.0 |

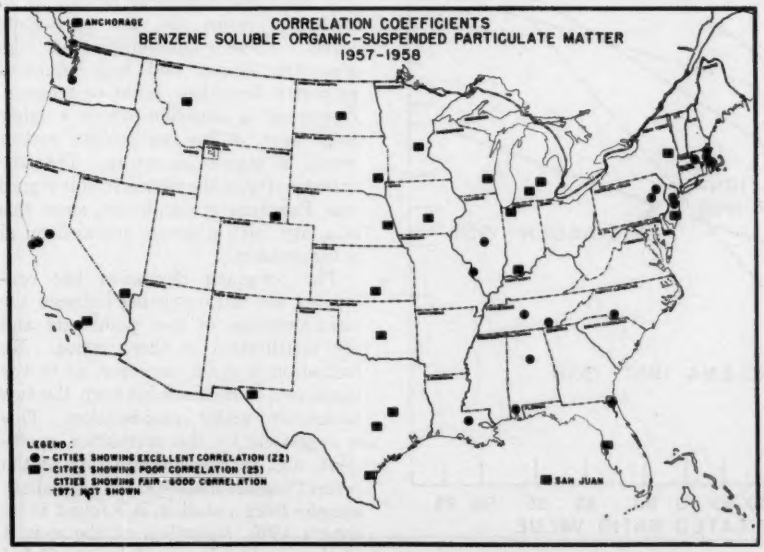


Fig. 4.

that the correlation was perfect and would have a value of one. On the other hand, if it were found that the percent of organic matter varied greatly for samples of the same weight and small samples showed low organic content, and large samples had a high organic content, the correlation would be poor and the coefficient would approach zero or be negative.

When correlation coefficients are computed on the basis of a relatively small number of samples, it must be recognized that the range of values represented by a given correlation coefficient is quite broad. Those ranges are listed in Table V for confidence limits at the 95% level; i.e., only one time in 20 would a value fall outside these limits by chance alone. For the purpose of the following discussion, it is stipulated that for correlation coefficients of less than 0.500, the correlation is poor; for 0.500–0.749, the correlation is fair; for 0.750–0.849, it is good; and for 0.850 and above, correlation is excellent. Of the 144 cities for which we have data, correlations were excellent for 22, good for 35, fair for 62, and poor for 25. The distribution of communities showing poor and excellent correlations between benzene soluble organics and suspended particulate matter is shown on the map (Fig. 4). Of the 25 communities showing the poorest correlation, 14 are located in the section of the United States where the occurrence of windblown dust is frequent. Most of the cities where the best correlation is found are generally industrial communities located in the South, or on the east or west coast.

There are 54 cities for which data are available for both 1957 and 1958. Satisfactory correlations were found between benzene soluble organic and suspended particulate levels for both years, as well as for the combined data in 35 of these cities. For 14 cities, the correlation was good for one year and poor for the other; when the combined data were considered, six of the cities demonstrated good correlation, while eight

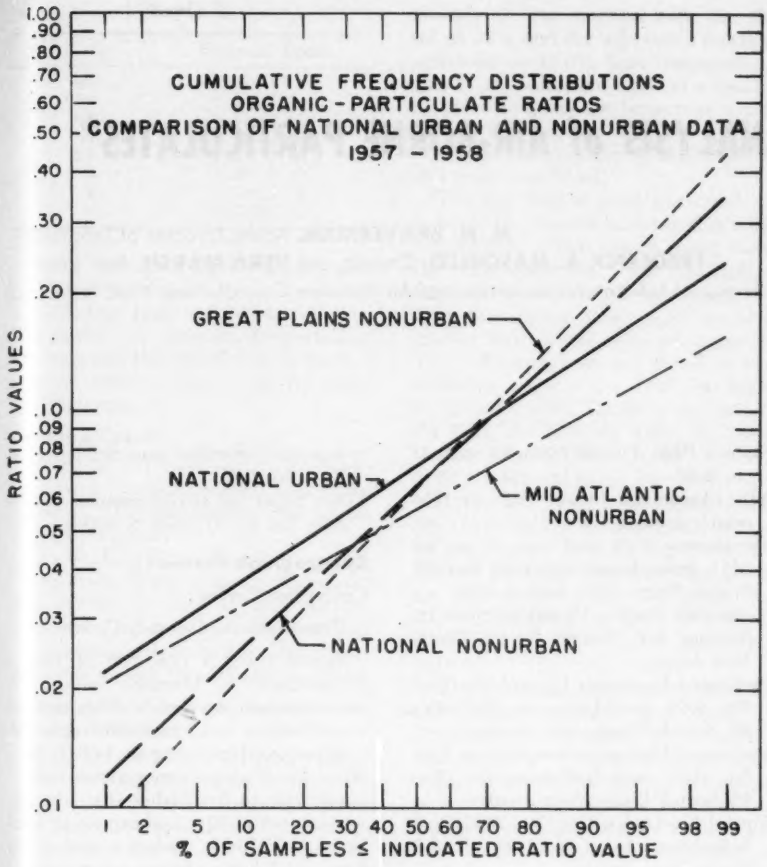


Fig. 5.

remained poor. No change was brought about when the two-year data for four cities showing poor correlations for the individual years were combined. In one case (Omaha) where satisfactory correlation was found for the individual years, when the combined data were used, the correlation became poor. On the basis of the foregoing it appears that, with a few exceptions, more reliable results are obtained when data for more than one year are combined.

To determine their full significance, organic to particulate ratios must be considered in connection with particulate levels, because for those stations that show high degrees of correlation, it is the particulate level as well as the organic to particulate ratio that determines the total concentration of the crude organic fraction in the air. If we take a look at the particulate levels of those cities for which the mean organic/particulate ratio is high, and at the organic/particulate ratios for those cities for which the mean particulate levels are high (Table VI), we can readily observe that it is necessary to consider both parameters to fully understand the situation.

**Table VII—Comparison of Urban and Nonurban Data Ratios—1957–58**

| | Benzene Soluble Organic to Suspended Particulate Ratios | | | | | |
|---|---|---|---|---|---|---|
| | | | | | Middle 80% of Values | |
| | No. of Stations | AM | GM | SGD | 10%–90% | 90%/-10% |
| Urban—Nat'l | 144 | 0.082 | 0.074 | 1.62 | 0.038–0.140 | 3.7 |
| Nonurban—Nat'l | 38 | 0.084 | 0.066 | 1.98 | 0.026–0.155 | 6.0 |
| Nonurban by Regions | | | | | | |
| New England | 3 | 0.074 | 0.067 | 1.83 | 0.028–0.140 | 5.0 |
| Mid Atlantic | 3 | 0.064 | 0.058 | 1.50 | 0.033–0.100 | 3.0 |
| Mid East | 4 | 0.084 | 0.074 | 1.73 | 0.034–0.155 | 4.6 |
| South East | 5 | 0.091 | 0.073 | 1.78 | 0.034–0.165 | 4.8 |
| Mid West | 5 | 0.061 | 0.052 | 1.73 | 0.025–0.105 | 4.2 |
| Great Plains | 6 | 0.088 | 0.066 | 2.25 | 0.022–0.190 | 8.6 |
| Gulf South | 1 | 0.056 | 0.050 | 1.63 | 0.024–0.100 | 4.2 |
| Rocky Mountain[a] | 4 | 0.117 | — | — | — | — |
| Pacific Coast | 6 | 0.093 | 0.073 | 2.15 | 0.022–0.185 | 7.4 |

[a] Ratio values not log-normally distributed.

### Nonurban Relationships

The national nonurban arithmetic mean value for the organic-suspended particulate ratios was 0.084, with the mean values for the individual stations ranging from a minimum of 0.046 (Kansas) to a maximum of 0.144 (Colorado). The arithmetic mean values for the states are evenly divided above and below the national mean, indicating a normal distribution of state values. Both the arithmetic and geometric means for the nonurban stations are about the same as for the urban stations.

Comparisons of national and regional parameters are made in Table VII, and illustrated in Fig. 5. The Great Plains and Pacific Coast regions are outstanding in that they have high standard geometric deviations and wide ranges for the middle 80% of the ratio values. In seven of the eight regions (Gulf South excepted), the nonurban stations show higher standard geometric deviations and, consequently, the middle 80% of values cover wider ranges than the urban stations in the same regions. In 20 of the 27 states where the nonurban data were log-normally distributed, the standard geometric deviations for the nonurban stations were higher than those for the urban stations.

When the data were categorized topographically, i.e., as Coastal, Farmland, Forest, Desert, or Mountain, and the parameters evaluated, no great differences were observed when comparisons were made with the data in Table VII. While a greater range for the middle 80% of the data was noticeable, most of the topographical categories followed the same general pattern with the 90%/10% values ranging from 4.6 to 7.1. The outstanding deviation was shown by the Desert group with a value of 11.5, the highest for any single station or group of stations, considered in this paper. This extreme variation is no doubt due to large numbers of samples contaminated with windblown dust, as higher standard deviations were also observed for stations in the Great Plains where dust storms are common. Comparison on the basis of trajectory of the air before arriving at a sampling station, i.e., coastal stations versus inland stations indicates no significant differences in mean values for the ratios. This leads one to speculate whether

(*Continued on page 427*)

# *Spectrographic Trace* ANALYSIS *of* AIR-BORNE PARTICULATES*

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The analysis of air-borne particulates by the spectrograph has been demonstrated. For trace analysis these procedures have been restricted to large samples collected over extended periods. It seemed desirable to develop a method capable of analyzing small samples collected during brief sampling periods as especially exemplified by filter tape spot samples. This approach will now be presented. Fundamental techniques will be described so that specialized procedures can be developed according to special needs that may arise.

## Experimental Details

The procedures presented here were designed for use in processing minute traces of metals present in small quantities of particulate matter. Special emphasis has been directed to the analysis of spots of filter tape samples but attention is also given to samples collected on ashless filter paper by high volume samplers which have been operated for one hour only. Successful procedures have been developed for milligram quantities of sample containing microgram quantities of numerous metals. No transfer of sample from filter paper is necessary but a number of spots may be combined to increase sensitivity when indicated by low visible intensity. The fundamental approach of these procedures is the use of the ashed sample rather than an acid leach which frequently introduces errors due to incomplete solubility of the metals and to varying amount of impurities leached from the filtering medium.

## Apparatus and Chemicals

Bausch and Lomb 1.5 meter grating spectrograph 5A/mm second order with dc power source, dc arc stand and adjustable sector and mount spectrum viewing box and magnifier

Jarrell-Ash Comparator Microphotometer

Wig-LBug Vial Mixer

Staplex High Volume Sampler with 11 cm head

AISI Automatic Smoke Sampler, Research Appliance Co.

Polystyrene Vials and Caps, Cat. No. 3111, Spex Industries, Inc., Box 98, Scotch Plains, New Jersey

1/8 in. Ball Pestles, Plexiglass, Spex Industries, Inc., Box 98, Scotch Plains, New Jersey

Preformed Electrodes Upper 1/8 in. Cat. No. 4019, Spex Industries, Inc., Box 98, Scotch Plains, New Jersey

Preformed Electrodes Lower 1/8 in. Cat. No. 4020, Spex Industries, Inc., Box 98, Scotch Plains, New Jersey

Funnel Electrodes, Cat. No. 3003, Spex Industries, Inc., Box 98, Scotch Plains, New Jersey

G Standards for 43 elements in graphite, Spex Industries, Inc., Box 98, Scotch Plains, New Jersey

$G_1$—0.1%, Spex Industries, Inc., Box 98, Scotch Plains, New Jersey

$G_2$—0.01%, Spex Industries, Inc., Box 98, Scotch Plains, New Jersey

$G_3$—0.001%, Spex Industries, Inc., Box 98, Scotch Plains, New Jersey

$G_4$—0.0001%, Spex Industries, Inc., Box 98, Scotch Plains, New Jersey

Graphite Powder ultra purity, Spex Industries, Inc., Box 98, Scotch Plains, New Jersey

SA No. 1 Film 35 mm, Spex Industries, Inc., Box 98, Scotch Plains, New Jersey

Metallic Indium Pellets

Ashless Filter Paper for high volume sampler Schleicher and Schuell Fast Flow No. 2W

Filter Paper for smoke sampler Whatman No. 41 1 1/2 wide × 100 ft

## Spectrograph Curves

### Calibration Curves

*Transmission—Intensity Curves*

Before using a new roll of film a Transmission - Intensity calibration curve must be prepared. This method of calibration was found most suitable:

Set up a GE mercury arc bulb (120 v, 60 cycles, 35 amp, catalog number 3901) about four to five inches from the slit which is set at 32 γ and expose as indicated in Table I. Select a pair of Hg lines which have a transmission ratio of approximately 1.5 and read the percent T on the microphotometer and record as indicated on Table I which is an actual example of this operation.

A preliminary curve is now plotted on co-ordinate paper plotting the lighter lines on the vertical axis and darker lines on horizontal axis. Also add 100/100 point and 0/0 point. Take point 95 on horizontal axis and find value on vertical axis which is 84. Take 84 on horizontal axis and continue until the following tabulated series is obtained: 95, 84, 65, 40, 21, 9, 4, 1.

Since an approximate ratio of 1.5 existed between the two sets of lines assign a geometric series in descending order of transmission as outlined in Table II.

**Table I**

| Spectrum No. | Exposure Time, Sec | Transmission Lighter Line, % | Transmission Heavier Line, % | Average LL | Average HL |
|---|---|---|---|---|---|
| 1 | 10 | — | — | 87 | 66 |
| 2 | 20 | 81 | 65 | 80 | 63 |
| 3 | 30 | 63 | 41 | 65 | 43 |
| 4 | 40 | 58 | 32 | 60 | 34 |
| 5 | 80 | 29 | 13 | 29 | 13 |
| 6 | 10 | 87 | 66 | | |
| 7 | 20 | 78 | 60 | | |
| 8 | 30 | 66 | 44 | | |
| 9 | 40 | 61 | 36 | | |
| 10 | 80 | 28 | 13 | | |
| 11 | Brass 30 sec | | | | |

*Presented at the 54th Annual Meeting of APCA, Commodore Hotel, June 11–15, 1961, New York, New York.

**Table II**

| *T*, % | *Exposure Ratio* |
|---|---|
| 95 | $1.5^0$– 1 |
| 84 | $1.5^1$– 1.5 |
| 65 | $1.5^2$– 2.25 |
| 40 | $1.5^3$– 3.38 |
| 21 | $1.5^4$– 5.06 |
| 9 | $1.5^5$– 7.59 |
| 4 | $1.5^6$–11.39 |
| 1 | $1.5^7$–17.10 |

Using two cycle log/log graph, plot percent T on vertical axis, and exposure or intensity ratio on horizontal axis. This is the T–I curve for this particular emulsion and the film should be given a number which is placed on its corresponding curve.

### Working Curves

From the T-I curve above, the working curves for each element can now be prepared. Indicate the film number on each working curve.

The working curves described here are based on the "internal standard method" and indium is used as the internal standard. In addition, the standard "G" metal mixture of the Spex Company was used as the element standards.

### Preparation of Indium Graphite Standard Mixture

Make up an indium chloride solution (1 ml = 1 mg In) by dissolving pure metallic indium in HCl with the aid of heat and diluting with $H_2O$ to the desired concentration. Weigh out two grams of pure Graphite into a porcelain dish or casserole (do not use platinum) and add one ml of In $Cl_2$ solution. Add a few ml of alcohol and stir with glass rod until homogeneous. Dry in oven with frequent stirring until dry and free-flowing.

Make up the following standard mixtures as shown in Table II.

The standard mixtures are weighed out into small plastic vials supplied by Spex Manufacturing Company, a small plastic bead added, vial covered with cap and agitated in Wiggle-bug for one minute. The cavity of an 1/8 in. carbon electrode No. 4020 Spex is then filled with the mixture by means of a micro-spatula and a plastic funnel (Spex). The electrode is then inserted into the ignition holder and fired for 30 seconds at four amperes with the slit set at 32 γ and the adjustable rotating sector set at 12.5% light transmission. The 11 exposures are made in a similar manner except for the brass road which fired at two amp for 15 sec.

Develop for 2 1/2 min in D19 at 70°F and process as usual.

The dry film is then processed to indicate the various lines starting with copper 3247 which can be found from the brass spectrum. The qualitative analysis is performed by measuring from the copper lines or by use of a master film in the microphotometer. The following lines are noted in the following order: Cu 3247, **In 3256, In 3258, Cd 3261, Cu 3274,** Ag 3280.7, **Zn 3282,** Zn 3345, **Sr 3351,** Ag 3383, **Co 3453.5, V 3184,** Ca 3179.3, Mo 3170.3, **Mo 3132.6,** Be 3131, **Be 3130.4,** Al 3082, **Bi 3067.7, Ni 3050.8, In 3039.4,** Fe 30206, Mn 2949.2, Si 2881.5, Sn 2863.3, Mg 2852, **Cr 2835.6, Pb 2833, Sn 2840, Mn 2795, As 2780.2,** Fe 2755, **Ge 2651,** Be 2650.5, **Sb 2598,** Al 2575, C 2478.3, B 2496.8, **B 2497.7,** Co 2407, Be 2348.6.

The boldfaced elements are used for quantitative analysis and each is processed for percent T in microphotometer correcting for background.

By means of T-I curve, convert the percent T of each element to Intensity. For each spectrum set up a ratio of the metal to be determined and the internal standard using any one of three indium lines. Using two-cycle log/log paper set up a curve plotting micrograms on horizontal axis against ratio of intensity of element over ratio of intensity of indium on vertical axis. It is best to prepare a separate curve for each element; however curves of the different spectrum lines of indium may be on same graph. Correct for impurities if present as indicated in blank in Spectrum No. 1.

The sequence of operation for each spectrum is as follows: (*1*) read percent T of element and internal standard, (*2*) refer to T-I curve to convert percent T to intensity, (*3*) set up ratio of intensity of element over intensity of internal standard, and (*4*) plot ratio against micrograms of element for each spectrum.

**Table III**

| *Spectrum No.* | *Standard Spex Mixture, Mg* % | | | *Pure Graphite, Mg* | *Graphite Indium, Standard Mixture* | *Constituent, μg* |
|---|---|---|---|---|---|---|
| 1 | | | | 20 | 30 | 0 |
| 2 | 10 | 0.001 | ($G_3$) | 10 | 30 | 0.1 |
| 3 | 20 | 0.001 | ($G_3$) | 0 | 30 | 0.2 |
| 4 | 5 | 0.01 | ($G_2$) | 15 | 30 | 0.5 |
| 5 | 10 | 0.01 | ($G_2$) | 10 | 30 | 1.0 |
| 6 | 20 | 0.01 | ($G_2$) | 0 | 30 | 2.0 |
| 7 | 5 | 0.1 | ($G_1$) | 15 | 30 | 5.0 |
| 8 | 10 | 0.1 | ($G_1$) | 10 | 30 | 10.0 |
| 9 | 20 | 0.1 | ($G_1$) | 0 | 30 | 20.0 |
| 10 | 5 | 1.0 | | 15 | 30 | 50.0 |
| 11 | Brass | | | | | |

### Sampling and Ashing

#### *A.I.S.I. Automatic Smoke Samplers*

The key operation in the analysis of the particulate matter collected by the smoke sampler is the selection of the spots. For a routine analysis a 24-hr strip is selected. The spots are cut out, ashed, and weighed. A corresponding weight of filter paper surrounding the spots is also weighed and represents the blank.

For high concentration of particulate matter as indicated by the visible intensity one or two spots and the surrounding filter paper are separately ashed and weighed.

Note: Ash at 600°C or below.

#### *High Volume Sampler*

Ash the S & S filter paper containing the sample and repeat on unused filter paper, using two or three sheets, for the blank.

Note: Ash all filter paper at 600°C or below.

#### *Procedure*

Transfer ashed sample or blank to polystyrene vial. If weight is less than 20 mg, add sufficient pure graphite to bring total weight to 20 mg. If weight of ash is more than 20 mg, take a 20 mg aliquot. Add 30 mg of graphite indium standard mixture, drop in one or two 1/8 in. plexiglass balls, cap vial, and agitate for one minute in Wig-L-Bug. Repeat procedure as for preparation of working curves.

After obtaining the intensity ratios of each metal compared to the standard indium lines, find micrograms of each metal sought by consulting the working curves. Correct for aliquots, if taken.

Calculate micrograms per cubic meter for each metal by dividing micrograms by volume of air processed.

#### *Semi-Quantitative Procedure*

A rapid semi-quantitative procedure eliminating the necessity for Microcomparator and Calibration Curves has been developed by A. J. Mittledorf (*The Spex Speaker*, Vol. II, No. 2, April 1957—available from Spex Industries, Inc., Box 98, Scotch Plains, New Jersey). The four G standards are exposed on first four spectrums, the following seven are ashed samples mixed with pure graphite. Compare in view box and estimate micrograms of each metal.

#### *Results*

Approximately 1000 atmospheric samples have been analyzed for copper, zinc, cadmium, strontium, cobalt, vanadium, molybdenum, beryllium, bismuth, nickel, chromium, lead, tin, manganese, antimony; and boron's modification to include mercury and arsenic is being explored. The samples were collected

(*Continued on page 427*)

# ANALYSIS of SMOKE OBSERVATIONS at Ottawa, Canada*†

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During the period July 1956 to May 1957, two-hourly measurements of smoke density were recorded at two sites in the city of Ottawa. A standard A.I.S.I. filter-paper-tape sampler was used and spots were evaluated by light transmission measurement.

Given a set of observations such as these, what is the best method of analysis? It is the intention in this paper to consider the question in the hope that a useful format may be developed. Similar observations are being taken in other Canadian cities such as Vancouver, Winnipeg, and Montreal. The present paper is therefore considered as a study in methodology. It is designed to serve four purposes:

1. To develop a suitable method of interpretation of smoke observations when there are no concurrent meteorological observations other than those taken at an airport perhaps five to 10 miles distant. The transport and dispersion of smoke depends on the wind. Therefore, any serious attempt to understand smoke concentration patterns must take account of the climatology of the area. What interpretation can be made of pollution records when only limited meteorological data are available?
2. To compare simultaneous smoke observations from two different locations in the same city. If suitable techniques can be developed, then perhaps the method can be extended to cases where there are more than two observing sites.
3. To compare ratios of peak to mean concentrations. This concept is becoming of increasing interest in the literature.
4. To yield some indication of smoke concentrations in Ottawa. This aspect is incidental to the main purpose of the paper.

## Observing Sites

Figure 1 is a map of the central section of the city of Ottawa. The sampling stations were located on the roofs of a downtown office building—the War Service Records Building (50 ft), and a hospital for the chronically ill—St. Vincent's Hospital (75 ft). Hereafter, the two stations will be denoted WRB and SVH.

The WRB site was selected to serve

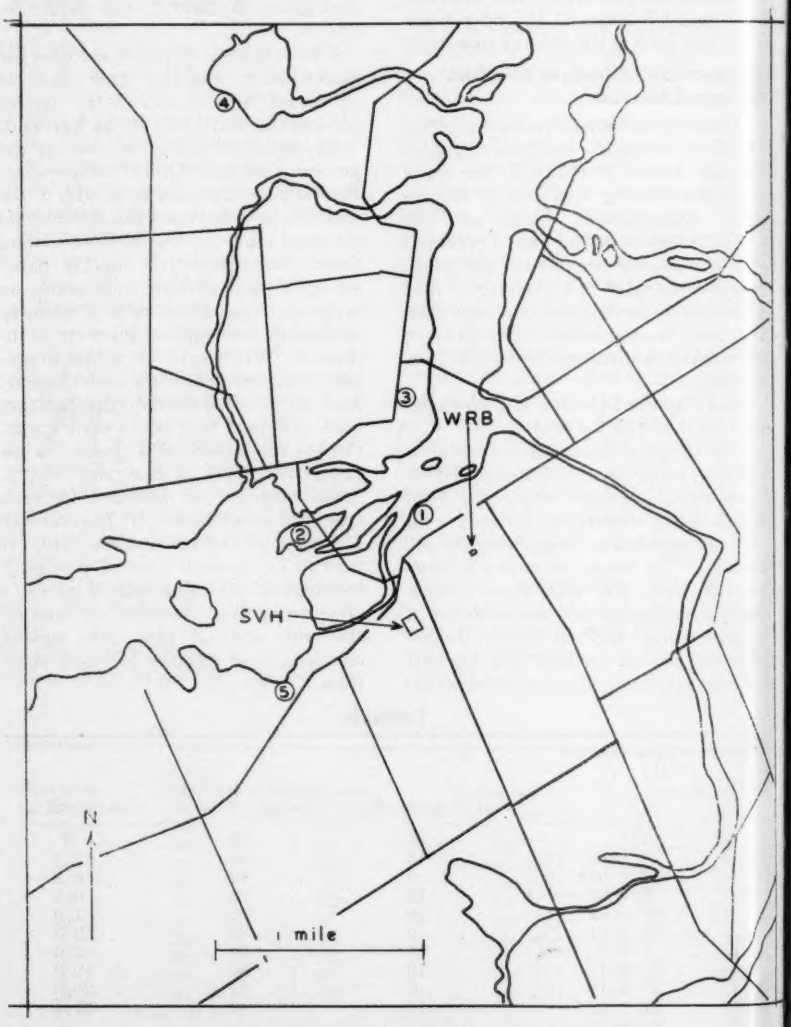


Fig. 1. Sampling sites and major point sources of pollution.

* Presented at the 54th Annual Meeting of APCA, Commodore Hotel, June 11–15, 1961, New York, New York.

† Published with the approval of the Director, Meteorological Branch, Toronto, and the Chief, Occupational Health Division, Department of National Health & Welfare, Ottawa.

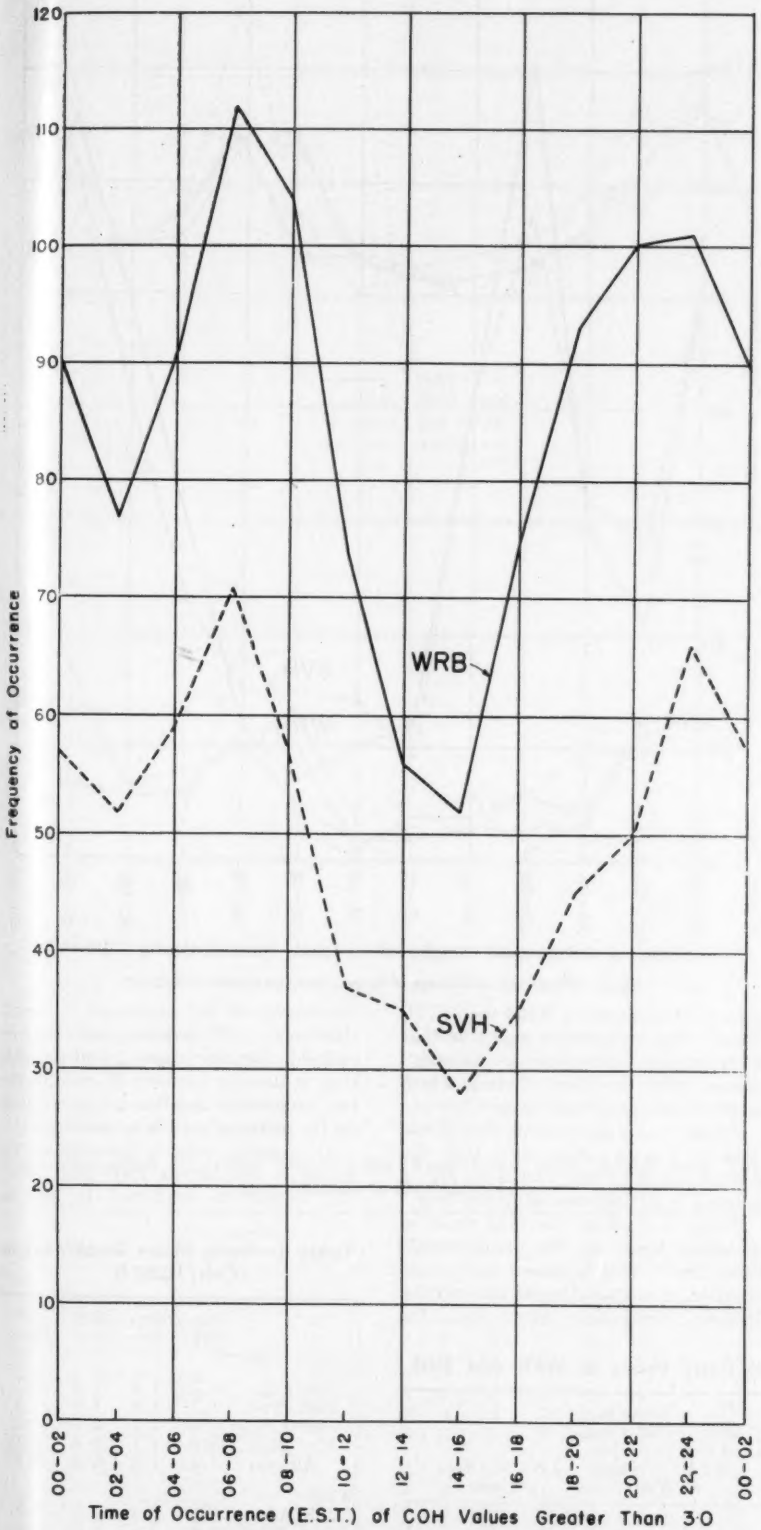


Fig. 2. Frequency distribution of Coh values greater than 3.0.

as a model air pollution sampling station. There is no stack in the immediate vicinity or on the building and it is in an area of moderate pollution. The SVH site was chosen because of a related epidemiological study, the results of which have not been published. At this site there was a stack of about 200 ft to the south on a separate central heating plant for the hospital. Stack height was about the same as the roof of the hospital. Summer emissions were at a much reduced rate—about 10% of those in winter.

Using the classification system outlined by Schumann and Gruber,[1] the area served by the sampler at SVH would be described as Zone BC (residential, high density, and commercial neighbourhood shopping district). The area served by the sampler at WRB would be described as Zone CB (commercial, central business district, and high density residential).

The topography of the area is relatively flat with a gentle slope down towards the Ottawa River. The climate is typically continental (January mean minimum temperature is 3°F above zero; July mean maximum is 80°F).

Hourly observations of wind are available from Ottawa Airport, which is about seven miles southsouthwest of the center of the city. Wind speeds over an open airport are likely to be stronger than over a built-up area; however, this bias is probably not too important in this study because rather broad class intervals have been used in tabulating smoke densities.

The other meteorological elements observed at the airport have not been used. For example, precipitation is known to have a cleansing effect on the atmosphere, but the interpretation of joint air pollution-precipitation records is difficult. A steady rain is usually followed by a frontal passage and wind shift. Therefore, it is not possible to tell whether the resulting improvement in air quality is due to the rain or to the fresh air mass and change in wind direction.

Another meteorological parameter that has been related to pollution levels is visibility (e.g. Leicester report[2]). However, the establishment of a joint association between visibility and smokiness would not contribute much to an understanding of the dispersion of pollution.

Major point sources of pollution that could have affected the two sampling sites are indicated on Fig. 1. They consist of a large central heating plant (*1*), paper mills (*2* and *3*), cement plant (*4*), and railway roundhouse (*5*) (no longer used).

Photographs of point sources are appended. St. Vincent's Hospital may be seen in the center of Fig. 9.

### Units of Measurement

The smoke densities are given in terms of Coh units per 1000 linear feet of air. Tebbens[3] believes that simple arithmetic means are adequate for summarizing accumulated data. However, Katz and Sanderson[4] suggest that the response of the A.I.S.I. sampler is not linear with increasing smokiness. This results in a variable bias of the data.

In the present study the emphasis has been placed on class frequency distributions rather than on mean values, in order to avoid the problem as much as possible.

### The Diurnal Variation of Smokiness

What is the best method for displaying a set of data in order to decide whether there is a diurnal cycle? Three possibilities are suggested:

1. Determine the mean value for each two-hourly sampling period. This technique will not be used for the reason advanced above.
2. Determine the number of cases in each two-hourly period when the Coh value was greater than some designated value. The data could be prestratified by initial sorting into seasons of the year.
3. Select the time of day of the highest Coh value from each day of record and determine the frequency distribution of these times.

The Ottawa data have been analyzed by methods (*b*) and (*c*), and the results are displayed in Figs. 2 and 3.

Figure 2 has the advantage that it shows the relative dirtiness of the air at the two stations. WRB evidently has poorer air quality than SVH. The disadvantage of Fig. 2 is that the data may contain many observations from a relatively few days when Coh values were high.

The method used in Fig. 3 ensures that only one observation will be taken from each day of record. Thus, a more representative picture is given of conditions throughout the entire year. This technique in one sense, therefore, "normalizes" the results, permitting comparison of sampling stations in different emission zones. For example, in Fig. 3 it is seen that the morning peak is more pronounced at WRB than SVH. Why? One explanation might be that SVH is more susceptible to "looping" plumes from the hospital stack which might occur at any time of day.

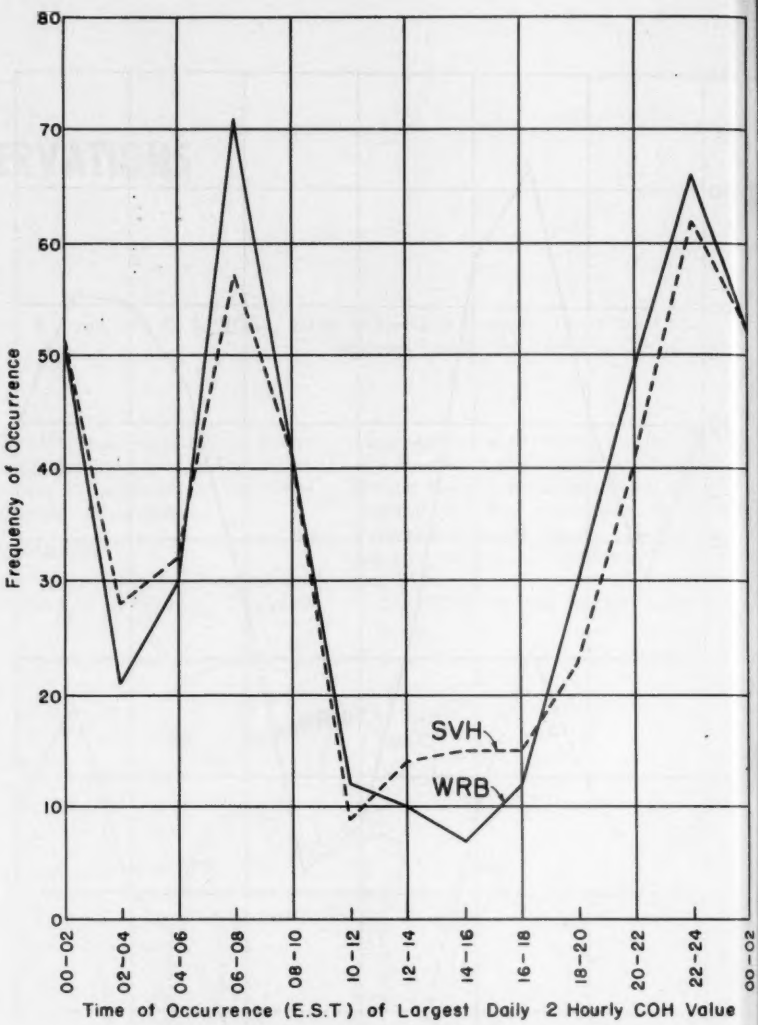


Fig. 3. Frequency distribution of largest daily two-hourly Coh value.

Figure 4 is a seasonal picture of the data in Fig. 3. It is seen that the general shape of the curves in Fig. 3 appears in all four seasons of the year.

There is a sharp morning peak in pollution levels in the period 0600–0800 EST. This is caused by a combination of increased emissions and the Hewson fumigation effect, i.e., the breakdown of the nocturnal inversion (Hewson[5]). The morning peak arrives earlier in the day in spring and summer than it does in winter. It may therefore be inferred that the primary cause for the morning peak is meteorological.

An evening peak in pollution levels occurs in the period 2200–2400 EST. This cannot be explained by an in-

**Table I—Frequency of Coincidence of Daily Peaks at WRB and SVH**

| | No. of Cases in Which SVH Daily Peak Coincides in Time with Peak at WRB | No. of Cases in Which SVH Peak of Coh Is Within ±2 Hr of Time at WRB | No. of Other Cases |
|---|---|---|---|
| July-Aug. | 13 | 20 | 30 |
| Sept.-Nov. | 21 | 14 | 51 |
| Dec.-Feb. | 30 | 17 | 36 |
| March-May | 33 | 19 | 39 |
| Year | 97 | 70 | 156 |

**Table II—Daily Mean Smokiness in Coh/1000 ft**

| | Sunday | Saturday | Mon.-Fri. | All Days |
|---|---|---|---|---|
| SVH | | | | |
| July-Aug. | 0.7 | 1.3 | 1.3 | 1.2 |
| Sept.-Nov. | 1.7 | 1.8 | 1.9 | 1.9 |
| Dec.-Feb. | 2.3 | 2.7 | 2.7 | 2.6 |
| Mar.-May | 0.8 | 1.1 | 1.4 | 1.2 |
| All year | 1.4 | 1.8 | 1.8 | 1.7 |
| WRB | | | | |
| July-Aug. | 0.7 | 0.8 | 1.0 | 1.0 |
| Sept.-Nov. | 1.8 | 2.1 | 2.3 | 2.3 |
| Dec.-Feb. | 3.0 | 3.4 | 3.2 | 3.3 |
| Mar.-May | 1.2 | 1.5 | 2.0 | 1.8 |
| All year | 1.7 | 2.0 | 2.2 | 2.2 |

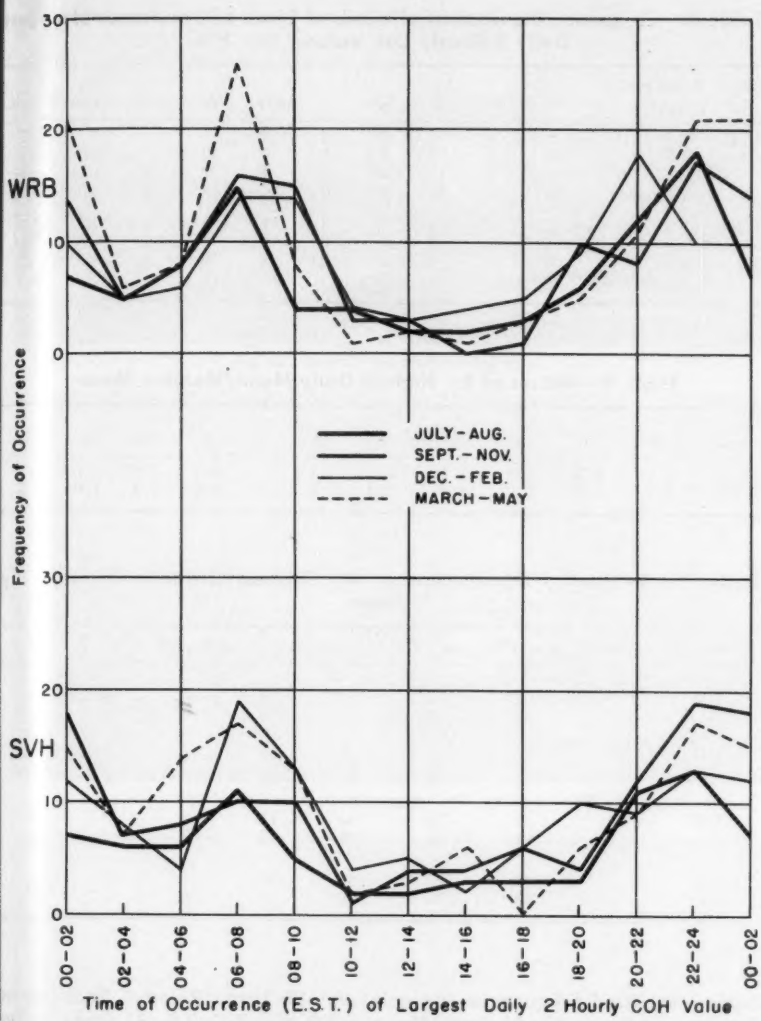


Fig. 4. Seasonal variations in frequency distribution of largest two-hourly Coh value.

crease in emissions but the exact meteorological mechanism is still obscure (Munn and Katz[6]).

Finally, it is noted that the trends at WRB and SVH are very similar. This is based on a statistical aggregate and it might be asked how frequently the times of daily peaks coincide at the two stations. The information is given in Table I.

**Table III—Wind Direction Frequencies Associated with Designated Coh Value Classes in Dec.-Jan.-Feb.**

| Station | Wind Speed, Mph | N | NE | E | SE | S | SW | W | NW | Calm | Totals |
|---|---|---|---|---|---|---|---|---|---|---|---|
| SVH | | | | | | | | | | | |
| (1) Coh ≥ 5.0 | 0–4 | 5 | 2 | 7 | 3 | 7 | 0 | 2 | 1 | 6 | 33 |
| | 5–9 | 3 | 4 | 10 | 0 | 17 | 6 | 0 | 0 | — | 40 |
| | ≥10 | 0 | 0 | 2 | 0 | 5 | 4 | 0 | 0 | — | 11 |
| | Totals | 8 | 6 | 19 | 3 | 29 | 10 | 2 | 1 | 6 | 84 |
| (2) Coh ≤ 0.7 | 0–4 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 |
| | 5–9 | 3 | 0 | 0 | 0 | 1 | 0 | 3 | 6 | — | 13 |
| | ≥10 | 2 | 0 | 0 | 0 | 0 | 0 | 20 | 17 | — | 39 |
| | Totals | 5 | 0 | 0 | 0 | 1 | 0 | 23 | 23 | 0 | 52 |
| WRB | | | | | | | | | | | |
| (1) Coh ≥ 5.0 | 0–4 | 10 | 5 | 8 | 8 | 7 | 2 | 3 | 1 | 6 | 50 |
| | 5–9 | 4 | 5 | 17 | 3 | 19 | 11 | 10 | 1 | — | 70 |
| | ≥10 | 0 | 6 | 18 | 3 | 13 | 8 | 4 | 1 | — | 53 |
| | Totals | 14 | 16 | 43 | 14 | 39 | 21 | 17 | 3 | 6 | 173 |
| (2) Coh ≤ 0.7 | 0–4 | 1 | 0 | 1 | 2 | 1 | 2 | 0 | 2 | 0 | 9 |
| | 5–9 | 1 | 1 | 1 | 1 | 7 | 2 | 0 | 4 | 0 | 17 |
| | ≥10 | 2 | 0 | 0 | 0 | 3 | 0 | 3 | 3 | — | 11 |
| | Totals | 4 | 1 | 2 | 3 | 11 | 4 | 3 | 9 | 0 | 37 |

It is seen that the peaks at the two stations occur simultaneously on 97 of the 323 days of the sample.

### The Weekly Cycle

Although it was stated previously that Coh values are not linearly related to smokiness, it is nevertheless true that a darker stain on the sampling tape will always be associated with a higher Coh value. Thus, an average daily Coh value has qualitative, although not quantitative meaning.

In Table II the daily means of smoke concentrations at the two stations have been tabulated separately for Sundays, Saturdays, and Mondays to Fridays. It will be observed that the Sunday average is consistently less than that for the Monday to Friday period at all times of the year. The two stations differ somewhat in absolute values but the percent variation between Sunday and Monday to Friday averages is quite uniform.

### The Relation Between Smokiness and Wind

In this study the Ottawa Airport wind associated with each two-hourly Coh value is the wind occurring at the midpoint of the sampling period. In any specific case this wind may be representative of the mean two-hourly wind. However, for a large statistical sample, the deviations should be random.

As in the case of the diurnal cycle, there are several ways of evaluating the relation between air quality and wind.

1 Determine the wind frequency distribution for all cases in which the Coh values exceed some designated value. A disadvantage is that the sample may then be weighted to favour a relatively few days when pollution levels remained high. This, perhaps, is a desirable feature; the object may be to determine what winds are associated with high pollution levels.

2 Select the highest Coh value from each day of record and determine the frequency distribution of the associated wind. This ensures data stratification, in that one wind observation from each day is included in the sample. However, it introduces a diurnal bias; it will be seen from Fig. 3 that a disproportionate number of observations will come from the periods 0600–0800 EST and 2200–2400 EST.

3 Select the wind observations to be used on the basis of peak/mean ratios. This will be considered in the following system.

In all three methods of analysis, a useful comparison can be made by determining wind frequency distributions when pollution levels are very low as

well as when they are very high. If the distributions are quite different, then it is an indication that wind is associated appreciably with pollution levels.

Table III presents the wind direction frequencies in December-January-February associated with Coh values greater than 4.9 and less than 0.8 respectively. It is necessary to consider one season separately to avoid bias; otherwise the high Coh values would be drawn largely from the winter months while the low values would be drawn mainly from the summer.

Table III shows that when air quality is poor, winds are primarily from the south, southwest, and east at both SVH and WRB. Very clean air is associated with west and northwest winds at SVH but there is no clear pattern at WRB.

The effect of wind speed on smokiness can be seen from the last column of Table III. For SVH, most cases of high Coh values are associated with light to moderate winds while occurrences of low Coh values are with strong winds. The WRB station, on the other hand, shows no clearly defined trend.

Table IV presents the wind direction frequencies in December-January-February at the times of occurrence of the largest daily two-hourly Coh values at SVH and WRB. As in the case of Table III, air quality is poor with winds from the south and east.

### Peak/Mean Ratios

The ratio $R_1$ will be defined as the ratio of the highest two-hourly Coh value in a day to the mean Coh value for the same day. Thus $R_1$ is a qualitative index of the strength of the diurnal influence on air quality.

The ratio $R_2$ will be defined as the ratio of the highest daily mean Coh value in a month to the mean Coh value for that same month. Thus $R_2$ is an index of the effect of broad synoptic-scale changes on day to day pollution loadings. The seasonal distribution of $R_2$ is given in Table V.

It is seen that the values of $R_2$ are quite similar at both stations in all seasons.

Consider next the ratios $R_1$. Their frequency distributions are given in Table VI.

It can be seen from Table VI that the values of $R_1$ are slightly higher at SVH than at WRB. This is due to the effect[1] of a neighbouring point source, the hospital central heating plant.

Table VI also shows that as the daily mean value of smokiness increases, the chances of the occurrence of a very large value of $R_1$ decreases.

Table VII has been prepared to display the association of wind with high values of $R_1$. The cases used were those in the upper tails of the

**Table IV—Frequency Distribution of Winds at Times of Occurrence of Largest Daily 2-Hourly Coh Values, Dec.-Feb.**

| Station | Wind Speed, Mph | N | NE | E | SE | S | SW | W | NW | Calm | Totals |
|---|---|---|---|---|---|---|---|---|---|---|---|
| SVH | 0–4 | 2 | 1 | 5 | 2 | 4 | 1 | 5 | 1 | 1 | 22 |
| | 5–9 | 6 | 4 | 7 | 0 | 9 | 8 | 9 | 1 | — | 44 |
| | ≥10 | 2 | 5 | 12 | 0 | 9 | 7 | 1 | 3 | — | 39 |
| | All speeds | 10 | 10 | 24 | 2 | 22 | 16 | 15 | 5 | 1 | 105 |
| WRB | 0–4 | 3 | 2 | 3 | 1 | 4 | 1 | 1 | 2 | 2 | 19 |
| | 5–9 | 4 | 6 | 3 | 0 | 14 | 5 | 7 | 1 | — | 40 |
| | ≥10 | 0 | 5 | 9 | 4 | 4 | 4 | 9 | 8 | — | 43 |
| | All speeds | 7 | 13 | 15 | 5 | 22 | 10 | 17 | 11 | 2 | 102 |

**Table V—Values of $R_2$: Highest Daily Mean/Monthly Mean**

| Station | J | A | S | O | N | D | J | F | M | A | M |
|---|---|---|---|---|---|---|---|---|---|---|---|
| SVH | 3.2 | 1.3 | 2.3 | 1.8 | 2.2 | 2.1 | 2.4 | 1.7 | 1.9 | 2.6 | 1.5 |
| WRB | 1.7 | 1.6 | 2.2 | 1.7 | 2.0 | 1.8 | 2.0 | 1.7 | 1.5 | 1.9 | 1.5 |

**Table VI—Frequency Distribution of $R_1$; Highest 2-Hourly Value/Daily Mean**

| Peak/Mean Ratio | 1.0–1.3 | 1.4–1.7 | 1.8–2.1 | 2.2–2.5 | 2.6–2.9 | 3.0–3.3 | >3.3 |
|---|---|---|---|---|---|---|---|
| Case 1: mean <1.1 | | | | | | | |
| WRB | 1 | 14 | 21 | 9 | 19 | 8 | 6 |
| SVH | 0 | 13 | 27 | 19 | 12 | 4 | 9 |
| Case 2: mean 1.1–1.9 | | | | | | | |
| WRB | 4 | 27 | 33 | 18 | 10 | 6 | 0 |
| SVH | 4 | 41 | 51 | 20 | 6 | 7 | 3 |
| Case 3: mean >1.9 | | | | | | | |
| WRB | 25 | 74 | 35 | 12 | 2 | 3 | 0 |
| SVH | 18 | 44 | 36 | 10 | 2 | 2 | 0 |
| All cases | | | | | | | |
| WRB | 30 | 115 | 89 | 39 | 31 | 17 | 6 |
| SVH | 22 | 98 | 108 | 49 | 20 | 13 | 12 |

**Table VII—Wind Frequencies Associated with Large Relative Peak/Mean Ratios; i.e. Cases of (A) Daily Mean ≤1.0 and $R_1$ >2.9 (B) Daily Mean 1.1–1.9 and $R_1$ >2.5 to (C) Daily Mean >1.9 and $R_1$ >2.1**

| Wind at the Hour of the Peak Coh Value | N | NE | E | SE | S | SW | W | NW | C |
|---|---|---|---|---|---|---|---|---|---|
| 0–4 mph | | | | | | | | | |
| WRB | 1 | 0 | 1 | 1 | 2 | 4 | 1 | 1 | 3 |
| SVH | 2 | 0 | 0 | 1 | 1 | 1 | 1 | 2 | 1 |
| 5–9 mph | | | | | | | | | |
| WRB | 0 | 0 | 2 | 6 | 6 | 4 | 6 | 3 | — |
| SVH | 0 | 1 | 2 | 1 | 7 | 10 | 4 | 1 | — |
| ≥10 mph | | | | | | | | | |
| WRB | 1 | 1 | 0 | 0 | 0 | 2 | 1 | 1 | — |
| SVH | 0 | 0 | 0 | 1 | 2 | 2 | 1 | 2 | — |
| All speeds | | | | | | | | | |
| WRB | 2 | 1 | 3 | 7 | 8 | 10 | 8 | 5 | 3 |
| SVH | 2 | 1 | 2 | 3 | 10 | 13 | 6 | 5 | 1 |

**Table VIII—Wind Direction Frequencies When Peak/Mean Ratios Are Large**

| | | N | NE | E | SE | S | SW | W | NW | C |
|---|---|---|---|---|---|---|---|---|---|---|
| Coh | WRB | 2 | 1 | 3 | 7 | 8 | 10 | 8 | 5 | 3 |
| | SVH | 2 | 1 | 2 | 3 | 10 | 13 | 6 | 5 | 1 |
| $SO_2$ | WRB | 9 | 0 | 4 | 2 | 6 | 2 | 8 | 30 | 3 |
| | SVH | 17 | 8 | 2 | 2 | 6 | 8 | 5 | 9 | 3 |

Fig. 5. Source 1—central heating plant.

Fig. 6. Source 2, at right—paper mill; source 1 at left.

Fig. 7. Source 3—paper mill.

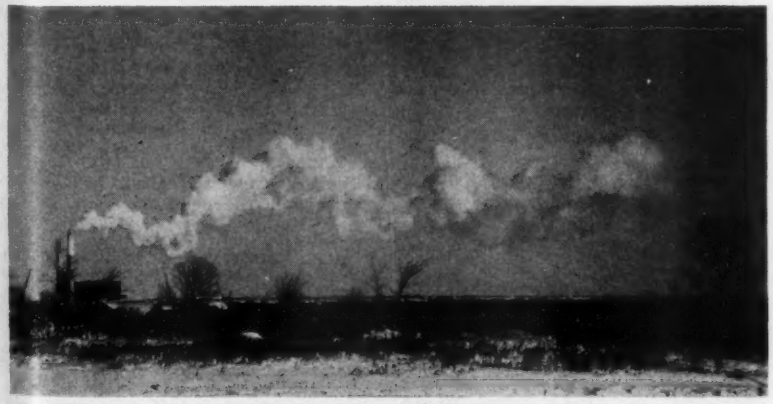

Fig. 8. Source 4—cement plant.

distributions in Table VI. This is why the designated class limits are different in the three cases.

The wind frequencies in Table VII are much like those in Tables III and IV except that the high proportion of east winds is not present. Thus, when pollution levels are high, they tend to be more uniform throughout the day with east than with southwest winds.

Tables III, IV, and VII represent three different methods of evaluating the relation between poor air quality and wind. From the standpoint of the air pollution control officer, the data presentation technique illustrated by Table III would appear to be the most direct, provided that care is taken to consider each season separately. However, Tables IV and VII may be indirectly helpful in interpreting results.

## Comparison with $SO_2$ Data

In a forthcoming paper a similar analysis of peak/mean ratios for sulfur dioxide at the same sites and for the same period of record will be considered. However, at this stage it is of interest to compare the last two lines of Table VII with similar data for $SO_2$. The comparison is given in Table VIII.

In the case of smoke densities, the highest peak/mean ratios are associated with south and southwest winds. For sulfur dioxide, on the other hand, the highest values are with northwest and north winds. Since the periods of record are identical, the difference cannot be attributed to meteorological factors. It therefore follows that point sources to the south and southwest produce more smoke and less $SO_2$ than point sources to the northwest and north. No detailed source inventory has been undertaken; nevertheless, the above conclusion seems reasonable to the authors. At the time of the survey an important smoke source to the southwest was a steam locomotive roundhouse and associated railway yard. There are also two lumberyards where wood shavings are burned in the area (Fig. 1).

For the Coh data in Table VIII there does not appear to be any important difference between the WRB and SVH sampling sites. However, in the case of $SO_2$, this is not so. Peak frequencies occur with northwest winds at WRB but with north winds at SVH. It is interesting (and perhaps significant) to note that a line drawn northward from SVH intersects a line drawn northwestward from WRB at the central heating plant (Source 1, Fig. 1).

## Changes in Time Scales

$R_1$ was the ratio of the highest two-hourly Coh value in a day to the mean Coh value for that same day. About

**Table IX—Extreme Values of $R_1$, $R_2$, and $R_3$**

| | No. of Values in Sample | Maximum Value of R WRB | SVH |
|---|---|---|---|
| $R_1$ | 320 | 4.0 | 7.0 |
| $R_2$ | 11 | 2.2 | 3.2 |
| $R_3$ | 1 | 1.5 | 2.6 |

320 different values of $R_1$ were thus obtained for each station, distributed according to Table VI.

$R_2$ was the ratio of the highest daily mean Coh value in the month to the mean Coh value for that same month. Eleven different values of $R_2$ were thus obtained for each station (Table V).

Let $R_3$ be the ratio of the highest monthly mean Coh value to the mean Coh value for the total 11-month sample. One value of $R_3$ is therefore available.

The extreme values of $R_1$, $R_2$, and $R_3$ are given in Table IX.

The decreasing value of R from $R_1$ to $R_3$ reflects the decreasing size to the statistical sample.

Gifford[7] and Singer[8] have given estimates of peak/mean ratios from a single continuous point source. Their averaging times range from three to 140 minutes, with peak times accordingly reduced, in contrast to the time scales considered here. Furthermore, as pointed out by Singer,[8] peak/mean ratios are strongly dependent on cross-wind distance; the ratios increase as the sampling site is removed away from the mean centre-line of the plume.

Gifford[7] estimates peak/mean ratios in terms of $t_A/t_p$, where $t_A$ is the averaging time of the mean and $t_p$ is the averaging time of the peak. For the data in Table IX, values of $t_A/t_p$ are 12, 30, and 12 for $R_1$, $R_2$, $R_3$ respectively. Examination of Gifford's Fig. 1 shows that our values of $R_1$, $R_2$, and $R_3$ fall between Gifford's estimated lines for surface and elevated sources. It is encouraging to find agreement in order-of-magnitude for such widely different scales of application.

## Conclusion

Simultaneous A.I.S.I. smoke observations made at two locations in central Ottawa have been analyzed in a number of different ways, along with wind speeds and directions taken at Ottawa Airport. It is believed that some of the techniques of data presentation and interpretation will be helpful to air pollution control officers.

Three ways have been suggested for display of the diurnal cycle of pollution:

1 Compute the mean value for each two-hourly period.
2 Count the number of cases in each two-hour period when the Coh values exceeded predesignated values (Fig. 2).
3 Select the time of day of the highest Coh value from each day and determine the frequency distribution of these times (Fig. 3).

Fig. 9. SVH site at center of photo. Source 5 was adjacent to left foreground.

Method *3* is preferred because it "normalizes" and stratifies the data, permitting comparison of sampling stations having widely different mean smokiness levels. A seasonal display is also desirable in order to ascertain whether the peaks show any time shifts from winter to summer (Fig. 4).

The weekly cycle in pollution has been examined by conventional methods (Table II). Since only one day in seven is a Sunday, more sophisticated techniques would require a much larger sample of data than was available in this study.

Three ways have been suggested for examining the relation of smokiness to wind:

1 Compare wind roses associated with very high Coh values with wind roses associated with very low Coh values (Table III).
2 Select the highest Coh value for each day and determine the frequency distribution of the associated wind (Table IV).
3 Determine the wind frequencies associated with high peak/mean ratios of smokiness (Table VII).

Method *1* is the most useful display for the air pollution control officer, provided that care is taken to consider each season separately in order to avoid bias. However, methods *2* and *3* may be indirectly helpful in interpreting results.

The effect of wind speed on pollution levels has been examined in the conventional fashion (Table IV).

A comparison of Coh values with $SO_2$ readings for the same sites and periods of records (Table VIII) can be particularly informative. For our data, the wind directions associated with high values are quite different for the two types of pollution.

Finally, a preliminary examination of peak/mean ratios has been undertaken (Tables V, VI, IX). On the average (Table VI), the two-hourly daily peak Coh value is about twice as large as the mean value for the day. The results are in order-of-magnitude agreement with estimates for peak/mean ratios by Gifford and Singer for single continuous point sources with sampling times on a much smaller scale. This study will be continued in a forthcoming paper on $SO_2$ data from the same sampling stations and period of operation.


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(*Continued on page 443*)

# INSTRUMENTATION for OLEFIN ANALYSIS At Ambient Concentrations*

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Few methods are known for the trace level determination of olefins in gas samples. Mader[1] described an extraction-spectrophotometric procedure which measures $C_4$ and higher molecular weight olefins at concentrations which exist in automobile exhaust; the method was not intended for the measurement of very low olefin concentrations. Altshuller, et al.,[2] reported a sensitive color test which works well at low levels; but, olefins, $C_4$ and higher, respond differently according to molecular weight and structure. Gas chromatography and mass spectrometry preceded by a concentration step are useful for separating and identifying the components of hydrocarbon mixtures including olefins.

This paper describes a bromination and coulometric system and the instruments developed to measure olefins in automobile exhaust and in the atmosphere. The concentration of olefins in automobile exhaust considered here is over 100 ppm while that in the atmosphere is usually less than 4 ppm. The method is based on the observation that the olefins, excluding ethylene, can be quantitatively brominated by passing a gas sample through a solution of appropriate composition. The method is based also on the principles of coulometric titration. The system differs from the conventional coulometric practice in the bromination is carried out before coulometry. The excess of bromine to do this is about $2 \times 10^{-5}$ moles per liter. For this reason, the system has been termed "bromocoulometric."

In this system the gas sample is passed through the sensing solution and the time is measured to generate the bromine absorbed by the sample. The bromine generated is measured amperometrically, that is, by noting the current flowing through the solution when a constant potential of 220 or 230 mv is applied to the electrodes. The current is a linear function of bromine concentration over a range of one to more than 20 microamperes. The equivalents of bromine added to the solution and, hence, the bromine absorbed by the sample can be determined by Faraday's Law. However, in the laboratory experimental apparatus, a 0 to 10 mv recorder was used to monitor the current. Thus, the recorder was used to indicate relative bromine concentration and not to measure bromine quantitatively. The generator "on time" is the quantitative value. Figure 1 shows a typical recorder trace obtained with laboratory equipment. The vertical axis is an arbitrary scale which can be equivalent to a bromine indicator current of 0–2, 0–8, 0–20, etc., microamperes depending on the circuitry and sensitivity setting. The horizontal scale represents the time spent during the analysis. The first plateau gives the stable concentration when no sample is being added. A slight downward drift here is caused by the slow loss of bromine which seems to take place with most of the solutions used in the basic study. As soon as the sample flow starts, bromine is used up much faster as indicated by the recorder trace. When no further rapid decrease occurs, the generator is started; and bromine concentration is restored as indicated by the heavy black line. The time spent in generation is a measure of the olefins in the sample. It is possible to correct for the slight loss of bromine, not due to the sample, by generating only to an extrapolation of the initial plateau.

The MDC Models 810-1 and 811-1 Olefin Analysers are instruments designed for use in the laboratory or field. They are portable instruments for automatic and continuous olefin analysis of a gas sample or stream and are de-

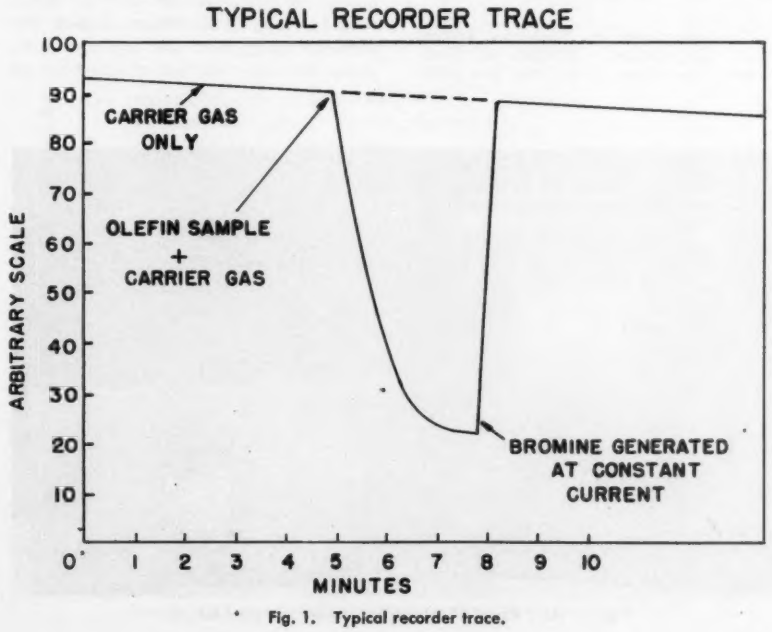


Fig. 1. Typical recorder trace.

* Presented at the 54th Annual Meeting of APCA, Commodore Hotel, June 11–15, 1961, New York, New York.

signed for operation with a Tally-Print Model BR* recorder modified slightly for this purpose. An analyser and print-out unit are shown in Fig. 2.

The essential parts of the Olefin Analyser are as follows:

1 Reaction chamber, motor driven
2 Meter-relay generating control system
3 Meter, generating current
4 Meter, sensing electrode voltage
5 Gas sample pump, constant volume type
6 Solution replenishment system
7 Dual power supply for generating and time-delay systems
8 Sensing electrode system with mercury battery
9 Tally-Print print-out recorder unit
10 Connecting cable attached to Tally-Print unit

Figures 2 and 3 show many of these parts.

The Model 810-1 is an Atmospheric Olefin Analyser. It has a 0 to 500 ua generating current meter and 150 or 500 ua generating current rates controlled by a toggle switch. The 150 ua rate is intended for olefin analysis in a range of 0 to 4 ppm (vol) and the 500 ua rate is intended for rapid increase of solution conductivity to a desired level or for olefin analysis in a range of approximately 0 to 25 ppm (vol).

The Model 811-1 is a High Concentration Olefin Analyser. It has a 0 to 10 ma generating current meter and a choice of nine or one ma generating current rates controlled by a toggle switch. The nine ma rate is intended for olefin analysis in a 0 to 1000 ppm (vol) range and the one ma rate is intended for olefin analysis in a range of approximately 0 to 100 ppm (vol).

* Manufactured by Standard Instrument Corporation, Division of Heat-Timer Corporation, New York, New York.

Both analysers have built in flexibility by the inclusion of the three meters and various adjustment controls. Necessary current and voltage adjustments are made easily in the field without need for any additional test instruments. Gas samples flow by a constant volume moving cylinder pump driven by a 300 rpm synchronous motor so that use of a flowmeter is not required.

If modification of the generating rates is desired, this can be accomplished by substituting a resistor of another value for one of the current limiting resistors which are mounted in an accessible location.

If modification of the fixed gas sample flow rate is desired, this can be accomplished by substituting another pump assembly of a desired capacity. By plugging the power cable from the print-out unit into a 115 v 60 cycle receptacle both the analyser and print-out unit are supplied the required external power. The analyser is connected to the print-out unit by a 25 foot cable. This allows for remote placement of the analyser where gas analysis is desired. In this way on-off switching and tape readouts are available at the recorder which can be located at a more convenient location.

## Reaction Chamber

The reaction chamber and its motor are mounted on the left side of the front panel. A 1.6 mfd capacitor is part of the motor circuit and is mounted on the rear surface of the chassis. The chamber contains a specific quantity of a bromide solution and two pairs of platinum electrodes. In operation the motor-driven rotating impeller draws the solution into its lower end, up its inner surface, and throws a continuous stream of solution out of four holes near its top. This stream strikes the deflector tube near the top and runs down the inner surface of this tube to the lower part of the chamber. Here the solution is available to repeat the rapid circulation over both electrode pairs and up into the impeller again. By means of one pair of electrodes bromine is generated in the solution until a desired conductivity level such as 10 ua is reached and controlled by the sensing electrodes and circuitry, at which time generation is stopped. The gas pump forces the sample into the top central part of the chamber by a path through an inlet tube, through the solution stream, and downward between the impeller and the deflector tube. Thus, the gas sample passes through the reaction chamber where it intimately contacts the sensing solution. Olefins present in the gas sample are brominated by contact with solution and thereby remove a definite amount of bromine. Electrical generation of bromine takes place intermittently as required to replace that used by the olefin reaction plus that lost by carry-out and decay.

As noted above, the sensing electrode pair measures the solution conductivity. This is used by the meter-relay control system to switch the generation current on and off as required to maintain the desired conductivity and bromine concentration level.

## Print-Out of Data

The generation on-time is representative of the olefin content of the gas sample. Generation on-time is recorded as a percentage by Tally-Print readout figures at five minute intervals. (This print-out interval can be changed if desired by substitution of a small motor with different gear ratio in the readout device.)

Interpretation of the print-out values in terms of olefin content in parts per million by volume requires the use of a graph. Figure 5 is such a graph. It contains curves plotted from results obtained by tests with the high concentration olefin analyser. These test results are representative of certain test conditions only and are useful for interpretation of print-out numbers of later analyses made under the same test

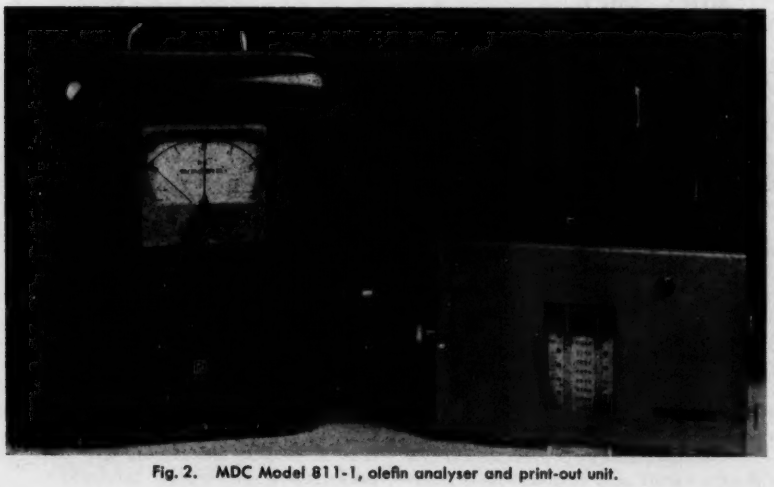

Fig. 2. MDC Model 811-1, olefin analyser and print-out unit.

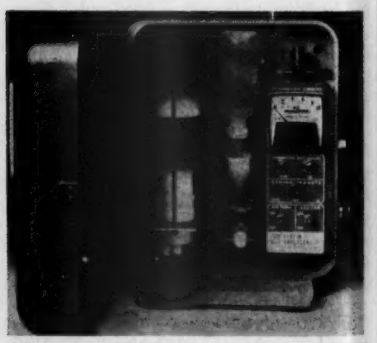

Fig. 3. Olefin analyser, inside front of MDC Model 811-1.

conditions which include: air flow rate, generating current rate, conductivity level, sensing electrode voltage, and solution composition.

As indicated in Fig. 5, only a brief generation on-time is required to replace bromine lost by sweep-out when the electrical generation rate is nine ma. Print-out data should average less than one percent for most indoor atmospheres under the tested conditions noted. This graph shows the results of tests made with olefin-air mixtures containing known quantities of one pure olefin gas diluted in room air. The gases, trans-2-butene, propylene, and ethylene, were sampled individually. This graph can be used to interpret print-out data obtained with olefin mixtures to a relative extent; that is, an analysis of an unknown olefin mixture that might contain several olefins can be reported in terms of an equivalent amount of one pure olefin such as propylene or trans-2-butene.

Certain variable conditions such as temperature of the solution can affect the test results. The preparation of additional graphs based on additional test conditions may be undertaken to improve the accuracy of print-out interpretations.

Figure 6 is a graph of print-out data of a Model 810-1 Atmospheric Olefin Analyser and applies to analyses made at the test conditions. Such variables as air and solution temperatures and air humidity affect the bromine carry-out characteristics of the gas sample stream. In the tests for Fig. 6 room air was used as a diluent. This air was not an ideal diluent because of its varying olefin content of less than one ppm.

### Solution Replenishment System

The efficiency of the reaction chamber is dependent upon the solution level in the chamber. There is a gradual loss of solution from the chamber due to carry-out by the gas sample stream. For sustained continuous operation it is desirable that solution be added in very small increments. The solution level is maintained at a constant level by the replenishment system when the reaction chamber motor is running. A suggested level is to have the top of the solution just touching the lower open edge of the glass deflector tube. The solution level in the chamber may be adjusted when desired by sliding the solution reservoir up or down in its mounting clamp.

The solution can be drained from the chamber, reservoir, or both simultaneously by turning the stopcock plug to a proper position.

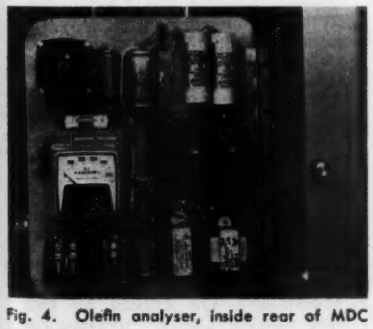

Fig. 4. Olefin analyser, inside rear of MDC Model 811-1.

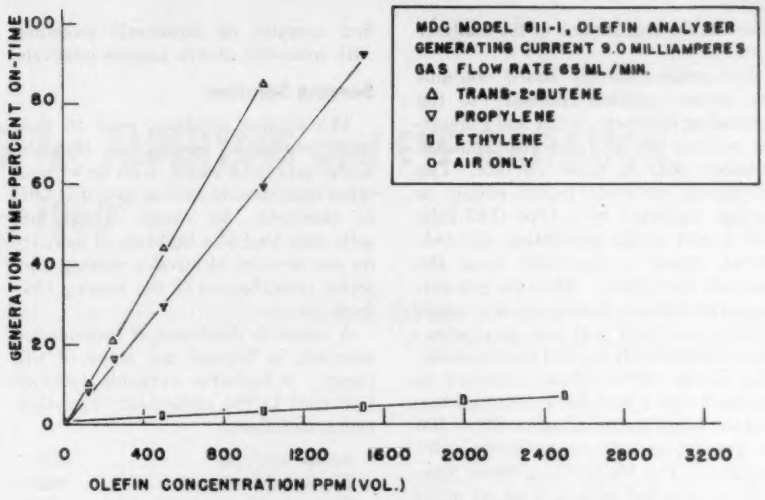


Fig. 5. Olefin concentration versus generation time for MDC Model 811-1.

### Gas Sample System

The gas sample system is designed to provide a constant flow rate. Essential components of the system are a constant-volume positive-displacement pump of a reciprocating cylinder type mounted in the upper right corner of the front panel. The pump has a 300 rpm synchronous motor with its associated one mfd capacitor mounted on the rear of the chassis behind the pump. The gas sample to be analyzed is drawn in through the gas sample tube and passes through the pump and reaction chamber, and then out the gas exhaust tube. An off-on switch for the gas pump motor is located on the front panel of the analyser unit.

One analyser has a 400 ml/min pump assembly and the other has a 65 ml/min capacity pump assembly.

### Electrical System

There are four electrical circuits in each analyser plus one in the read-out unit. The four electrical systems of the analyser unit are: the sensing electrode circuit, the ac power circuit, the generating electrode circuit, and the generating control circuit. Some components are common to two or more circuits.

The ac power circuit includes the motors of the pump and reaction chamber and the input to two transformers that are mounted on a printed circuit

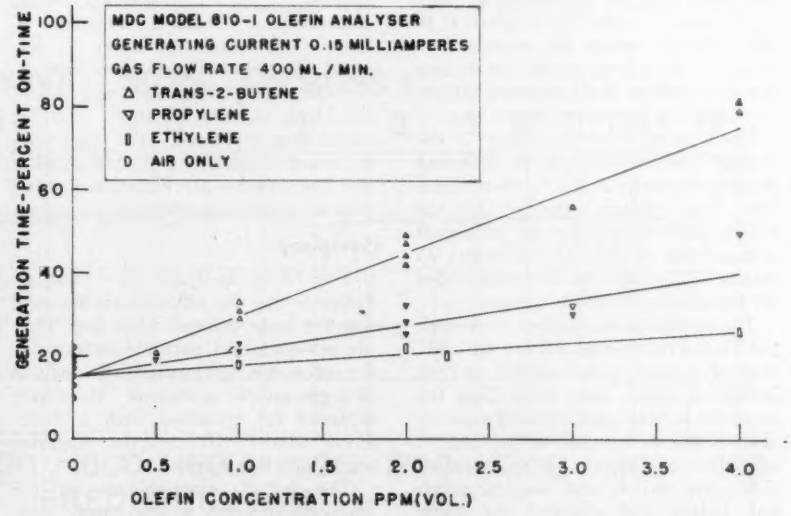


Fig. 6. Olefin concentration versus generation time for MDC Model 810-1.

board in the rear section of the analyser unit.

The printed circuit board contains two power supplies necessary to the generating electrode circuit and generating control circuit, and also contains resistors used in these circuits. The generating electrode power supply is voltage regulated by a type OA2 tube and is part of the generating electrode circuit which is controlled from the analyser front panel. Here the generating current meter, generating-rate toggle switch, and high and low generating-adjust controls are located conveniently. The Model 810-1 Olefin Analyser is supplied with a 0 to 500 microammeter, and the toggle switch allows a 150 or 500 ua generating rate or a center "off" position. The Model 811-1 Olefin Analyser is supplied with a 0 to 10 milliammeter, and the toggle switch allows a one or nine ma generating rate or a center "off" position. A platinum electrode pair in the reaction chamber completes the generating electrode circuit.

The generating control circuit is of the meter-relay type. It contains the unregulated power supply on the pc board, a meter-relay adjustable from 0 to 20 ua with a control knob providing a visual check and control of solution conductivity level, a meter-relay r-c delay system (capacitors, resistors, spdt relay) and a dpdt relay mounted in the rear of the analyser. The dpdt relay closes generating current and print-out switches simultaneously, so readout figures are a record of generating current on-time.

The sensing electrode circuit consists of the meter coil of the meter-relay, a toggle switch on the chassis front panel, and components on the chassis left rear plus a pair of platinum sensing electrodes in the reaction chamber.

The components on the chassis left rear are a 0 to 500 microammeter with appropriate circuitry to indicate 0 to 500 millivolts across the sensing electrodes, a fine adjust control for sensing electrode voltage, and a mercury battery in a clip above the meter chassis box.

The sensing electrode voltage is the voltage across the pair of platinum sensing electrodes in the reaction chamber. This voltage, supplied by the replaceable mercury battery, is sensed over a range of 0 to 500 millivolts. A setting of 230 millivolts is recommended for the sensing electrode voltage.

The modifications made to the original Tally-Print Model BR are the addition of a main power switch and an indicating panel light controlling the ac power to both print-out and analyser unit, change of fuse size to one ampere, and the elimination of a manual-automatic switch and manual print-out button not required for olefin analyzing. The Tally-Print as modified operates on automatic recording with print-out at five minute intervals.

### Sensing Solution

The sensing solutions used to date have contained potassium bromide, acetic acid, and water, with or without other constituents such as mercuric salts as catalysts. An excess of mercuric salts may lead to a build-up of mercury on one or more electrodes causing electronic disturbances in the sensing electrode circuit.

A complete discussion of bromination solutions is beyond the scope of this paper. A tentative workable composition used in the preliminary investigation is as follows:

| | |
|---|---|
| Acetic acid, ml | 450 |
| Methanol, ml | 950 |
| Water, ml | 150 |
| Potassium bromide, g | 16 |
| Mercuric acetate, g | 0.5 |

The addition of zinc sulfate and more water appeared to have a beneficial effect in promoting complete recovery of propylene, but others[4] have reported opposite results. The solution selected for the foregoing instruments eliminated the methanol and mercuric acetate.

The choice of solution composition may depend on the concentration range being measured and on the objectives in mind. Flow rate, temperature, and the presence of inert hydrocarbons in the sample may also have an effect. The final choice of solution will require a systematic study of the many possible combinations.

Pretreatment of the solution with an excess of bromine may prove to be desirable.


### Acknowledgments

The authors appreciate the guidance and assistance received from the following personnel of Mast Development Company, Inc.: Robert G. Kling, Experimental Chemist; Timothy B. Zimmerman, Engineer; Jerry V. Sergeant, Electrical Engineer; Samuel E. Hohulin, Design Engineer; and Gifford M. Mast, General Manager; and the model shop personnel. The encouragement and assistance of the personnel of the Los Angeles Air Pollution Control District are also appreciated.


### Summary


Two Olefin Analysers have been developed—one for atmospheric use and one for auto exhaust analysis. They are self-contained portable instruments for automatic, continuous olefin analysis of a gas sample or stream. These were designed for operation with a Tally-Print Model BR recorder modified slightly for this purpose.

The initial investigations on the bromocoulometric system were conducted in the laboratories of the Los Angeles Air Pollution Control District, 760 South Ferris Avenue, Los Angeles 22, California, and the instrument development by Mast Development Company, Inc., was accomplished under an Air Pollution Foundation contract.


## SECTION HI-LITES

### West Coast Section

Some 60 guests turned out for the June 29 Los Angeles Dinner Meeting of the West Coast Section. The meeting was held at the Roger Young Auditorium and the general topic for the evening was the recently concluded New York National Meeting.

John H. Fairweather, Chairman of the West Coast Section, served as emcee and reported on the more intimate details of the New York convention, at which he delivered a paper.

Newly elected APCA President and section member, Dr. Larry Faith, gave a detailed report of the business meetings conducted in New York and outlined his APCA aims for the coming year.

Highlight of the evening was Ralph L. Weimer's personalized account of the several technical papers delivered by West Coast Section members.

A dinner meeting was held August 11 at the Kings Inn, San Diego. The program included speakers Dr. Robert E. Holzer, Professor of Geophysics at UCLA, and Dr. Albert F. Bush, Professor of Sanitary Engineering at UCLA.

Dr. Holzer's talk was concerned with positive and negative ions in the atmosphere, their relationships with air pollution and the physiological effects. He dealt with methods of detecting and measuring atmospheric ions, and observations on their occurrence and distribution in the atmosphere.

Dr. Bush had unusual photomicrographs illustrating his lecture which was most enlightening on the nature of atmospheric particles.

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3. Stephen W. Nicksic, "Instrumentation for Olefin Analysis at Ambient Concentrations," Fourth Conference on Methods in Air Pollution Studies, Los Angeles, California (Dec. 5–6, 1960).
4. Marcel Eye, Los Angeles Air Pollution Control District (private communication).

# *Atmospheric* SAMPLING *for* ALDEHYDES *and* EYE IRRITATION *in Los Angeles Smog—1960**,†

**N. A. RENZETTI,** Ph.D.,‡ Air Pollution Foundation, San Marino, California, and **R. J. BRYAN,** Los Angeles County Air Pollution Control District, Los Angeles, California

During the past several years we have produced eye irritating mixtures by irradiating either auto exhaust or single olefinic hydrocarbon-nitrogen oxides mixtures, in pure air systems.[1, 2] These results have been confirmed by other investigators.[3] One outcome of the work at the Stanford Research Institute Laboratories is the development of a model which can rationalize the observed eye irritation with the experimentally determined irritation from formaldehyde and acrolein.[4] Stephens added another feature to this model by the inclusion of peroxyacetylnitrate as an eye irritant.[5] A preliminary examination of this model was reported by Renzetti and Schuck.[6] The purpose of the experimental program reported in this paper was to provide an intensive test of the model by extensive analyses of smog *in situ* for aldehyde concentrations with simultaneous observations on severity of eye irritation.

* Supported in part by National Institutes of Health Grant RG6828 to Stanford Research Institute, South Pasadena, California.

† Presented at the 54th Annual Meeting of APCA, Commodore Hotel, June 11–15, 1961, New York, New York.

‡ Presently Consultant, Stanford Research Institute, South Pasadena.

§ South Pasadena work carried out by Truesdail Laboratories under contract to the Air Pollution Foundation.

## Experimental Procedure

We procured and analyzed samples of the atmosphere at 434 South San Pedro Street (6th floor) five days a week at 9, 10, 11, 12 AM and at 820 Mission St., South Pasadena§ (street level) at 1, 2, 3, 4 PM from July 5 to Nov. 18, 1960. Sampling for total aldehydes and formaldehyde occupied a ten-minute period centered on the hour. For each of the eight sampling times during the day a 150 liter polyethylene bag sample of the atmosphere was obtained for subsequent (within $^1/_2$–3 hours) analysis in a dual cell long-path infrared spectrometer by E. Schuck of Stanford Research Institute for compound X (peroxyacylnitrate).

Eye irritation data were obtained by questionnaires completed by individual panel members within five minutes of the scheduled hour. The Los Angeles panel was composed of District employees in their quarters on the six floors at the South San Pedro location (not air conditioned). The South Pasadena panel was composed of 12 young male college students. This panel has been making observations at the Stanford irradiation facility for a number of years. They were also used for the study reported in Ref. 6, wherein it was indicated that this panel responded to eye irritation with fair correlation to that of the public at large. In the present study only one or two values a day were obtained. The 2 PM value was obtained by having each panel member record his response to smog wherever he happened to be in the Los Angeles Basin. The 4 PM value was obtained from the panel members' response at the South Pasadena aldehyde sampling site.

## Analytical Methods

*1* Sampling and analytical procedures for determination of total aldehydes and formaldehydes. The sampling apparatus consisted of three absorbers, each containing 10 ml of one percent sodium bisulfite solution, connected in series through vapor traps to a Sprague

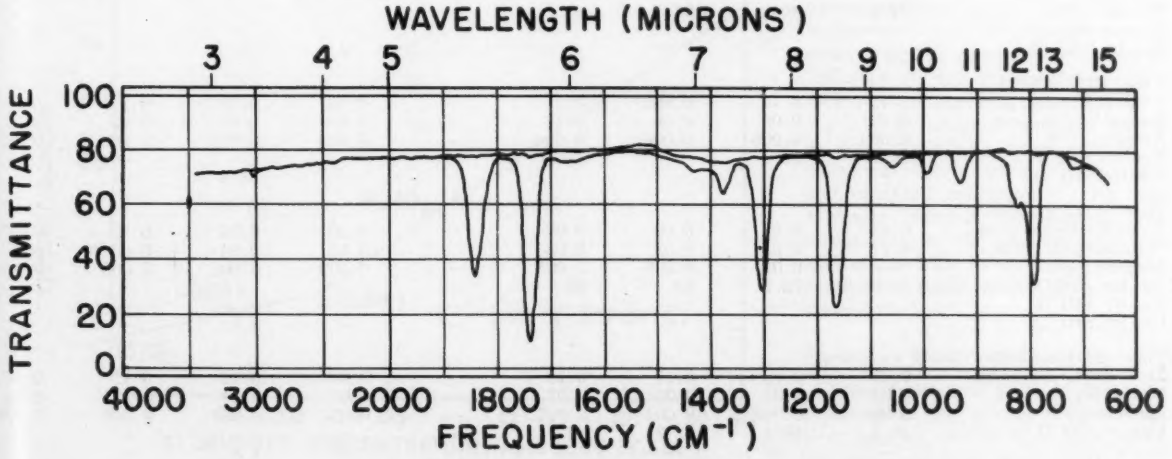


Fig. 1. Infra-red spectrum of compound X.

**Table I—Eye Irritation and Aldehyde Data**

| Location → | Downtown Los Angeles | | | | | South Pasadena | | | |
|---|---|---|---|---|---|---|---|---|---|
| Time: PDT 7/15/60 | 0900 | 1000 | 1100 | 1200 | 1200 | 1300 | 1400 | 1500 | 1600 |
| Total aldehydes, ppm | 0.28 | 0.33 | 0.25 | 0.16 | — | 0.15 | 0.10 | 0.04 | 0.04 |
| Formaldehyde, ppm | 0.09 | 0.07 | 0.08 | 0.04 | — | 0.05 | 0.04 | 0.03 | 0.02 |
| Eye irr. APCD % | 52.1 | 89.0 | 90.0 | 78.6 | — | — | — | — | — |
| Time: PDT 7/18/60 | | | | | | | | | |
| Total aldehydes, ppm | 0.40 | 0.25 | 0.06 | 0.16 | — | 0.12 | 0.11 | 0.12 | 0.14 |
| Formaldehyde, ppm | 0.01 | 0.02 | 0 | 0 | — | 0.04 | 0.03 | 0.04 | 0.05 |
| PAN, ppm | 0.05 | 0.03 | 0.02 | 0.02 | — | — | — | — | — |
| Eye irr. APCD % | 18.4 | 42.3 | 46.3 | 34.0 | — | — | — | — | — |
| Eye irr. SRI (1) | — | — | — | — | — | — | — | — | 5.0 |
| Eye irr. SRI (2) | — | — | — | — | — | — | 7.2 | — | — |
| Time: PDT 7/19/60 | | | | | | | | | |
| Total aldehydes, ppm | 0.21 | 0.14 | 0.16 | 0.17 | — | 0.23 | 0.13 | 0.11 | 0.07 |
| Formaldehyde, ppm | 0.03 | 0.0 | 0.03 | 0.03 | — | 0.07 | 0.05 | 0.04 | 0.04 |
| PAN, ppm | — | 0.05 | 0.01 | 0.01 | — | — | — | — | — |
| Eye irr. APCD % | 9.8 | 13.2 | 27.3 | 48.9 | — | — | — | — | — |
| Eye irr. SRI (2) | — | — | — | — | — | — | 11.4 | — | — |
| Time: PDT 7/22/60 | | | | | | | | | |
| Total aldehydes, ppm | 0.06 | 0.06 | 0.15 | 0.21 | — | 0.11 | 0.04 | 0.03 | 0.02 |
| Formaldehyde, ppm | 0.04 | 0.06 | 0.04 | 0.04 | — | 0.05 | 0.03 | 0.02 | 0.02 |
| Acrolein, ppm | — | — | — | — | — | 0.008 | 0.006 | 0.006 | 0.005 |
| PAN, ppm | 0.00 | 0.02 | 0.05 | 0.02 | — | — | — | — | — |
| Eye irr. APCD % | 6.2 | 42.0 | 63.3 | 53.8 | — | — | — | — | — |
| Eye irr. SRI (2) | — | — | — | — | — | — | 5.3 | — | — |
| Time: PDT 8/11/60 | | | | | | | | | |
| Total aldehydes, ppm | 0.08 | 0.16 | 0.11 | 0.07 | — | 0.09 | 0.05 | 0.02 | 0.01 |
| Formaldehyde, ppm | 0.05 | 0.07 | 0.04 | 0.03 | — | 0.03 | 0.01 | 0.01 | 0.01 |
| Acrolein, ppm | — | — | — | — | — | 0.007 | 0.006 | 0.005 | 0.005 |
| PAN, ppm | 0.00 | 0.00 | 0.00 | 0.01 | — | — | — | — | — |
| Eye irr. APCD % | 17.5 | 50.6 | 59.0 | 41.5 | — | — | — | — | — |
| Eye irr. SRI (2) | — | — | — | — | — | — | 7.1 | — | — |
| Time: PDT 8/18/60 | | | | | | | | | |
| Total aldehydes, ppm | 0.19 | 0.25 | 0.11 | 0.14 | — | 0.17 | 0.12 | 0.07 | 0.12 |
| Formaldehyde, ppm | 0.04 | 0.04 | 0.04 | 0.04 | — | 0.06 | 0.03 | 0.01 | 0.01 |
| Acrolein, ppm | — | — | — | — | — | 0.011 | 0.007 | 0 006 | 0 005 |
| PAN, ppm | — | — | 0.04 | 0.07 | — | — | — | — | — |
| Eye irr. APCD % | 3.7 | 22.2 | 51.3 | 50.0 | — | — | — | — | — |
| Eye irr. SRI (2) | — | — | — | — | — | — | 7.1 | — | — |
| Time: PDT 9/6/60 | | | | | | | | | |
| Total aldehydes, ppm | 0.36 | 0.27 | 0.19 | 0.23 | — | 0.14 | 0.09 | 0.11 | 0.08 |
| Formaldehyde, ppm | 0 | 0.10 | 0.08 | 0.05 | — | 0.02 | 0.02 | 0.03 | 0.02 |
| Acrolein, ppm | 0.005 | 0.011 | — | 0.005 | — | 0.007 | 0.008 | 0.008 | 0.008 |
| PAN, ppm | 0 | 0 | 0 | 0.05 | — | — | — | — | — |
| Eye irr. APCD % | 27.8 | 70.0 | 84.4 | 71.7 | — | — | — | — | — |
| Eye irr. SRI (2) | — | — | — | — | — | — | 6.9 | — | — |
| Time: PDT 9/7/60 | | | | | | | | | |
| Total aldehydes, ppm | 0.02 | 0.12 | 0.08 | 0.13 | — | 0.18 | 0.11 | 0.11 | 0.07 |
| Formaldehyde, ppm | 0.02 | 0.08 | 0.03 | 0.03 | — | 0.04 | 0.03 | 0.03 | 0.03 |
| Acrolein, ppm | 0.003 | 0.004 | 0.004 | 0.004 | — | — | 0.008 | 0.008 | 0.008 |
| PAN, ppm | 0 | 0 | 0.02 | 0.04 | — | — | — | — | — |
| Eye irr. APCD % | 12.9 | 49.2 | 61.7 | 63.5 | — | — | — | — | — |
| Eye irr. SRI (1) | — | — | — | — | — | — | — | — | 5.3 |
| Eye irr. SRI (2) | — | — | — | — | — | — | 12.7 | — | — |
| Time: PDT 9/8/60 | | | | | | | | | |
| Total aldehydes, ppm | 0.13 | 0.15 | 0.08 | 0.21 | — | 0.16 | 0.18 | 0.11 | 0.07 |
| Formaldehyde, ppm | 0.13 | 0.08 | 0.08 | 0.12 | — | 0.04 | 0.03 | 0.02 | 0.02 |
| Acrolein, ppm | 0.009 | 0.006 | 0.006 | 0.004 | — | 0.008 | 0.010 | 0.008 | 0.008 |
| Eye irr. APCD % | 16.7 | 52.4 | 61.9 | 41.5 | — | — | — | — | — |
| Eye irr. SRI (2) | — | — | — | — | — | — | 14.4 | — | — |
| Time: PDT 9/12/60 | | | | | | | | | |
| Total aldehydes, ppm | 0.12 | 0.05 | 0.09 | 0.09 | — | 0.20 | 0.16 | 0.10 | 0.10 |
| Formaldehyde, ppm | 0.11 | 0.05 | 0.08 | 0.08 | — | 0.03 | 0.04 | 0.02 | 0.02 |
| Acrolein, ppm | — | 0.011 | 0.006 | 0.005 | — | 0.009 | 0.008 | 0.006 | 0.008 |
| Eye irr. APCD % | 63.0 | 72.2 | 82.4 | 82.7 | — | — | — | — | — |
| Eye irr. SRI (1) | — | — | — | — | — | — | — | — | 1.0 |
| Eye irr. SRI (2) | — | — | — | — | — | — | 8.0 | — | — |
| Time: PDT 9/13/60 | | | | | | | | | |
| Total aldehydes, ppm | 0.05 | 0.08 | 0.17 | 0.13 | — | 0.09 | 0.12 | 0.12 | 0.13 |
| Formaldehyde, ppm | 0.02 | 0.07 | 0.03 | 0.07 | — | 0.03 | 0.04 | 0.03 | 0.03 |
| Acrolein, ppm | 0.003 | 0.004 | 0.004 | 0.005 | — | 0.007 | 0.009 | 0.008 | 0.009 |
| Eye irr. APCD % | 19.4 | 46.4 | 52.2 | 44.9 | — | — | — | — | — |
| Eye irr. SRI (2) | — | — | — | — | — | — | 4.8 | — | — |

*(Continued on following page)*

**Table I (Continued)**

| Location → | Downtown Los Angeles | | | | | South Pasadena | | | |
|---|---|---|---|---|---|---|---|---|---|
| Time: PDT 9/26/60 | 0900 | 1000 | 1100 | 1200 | 1200 | 1300 | 1400 | 1500 | 1600 |
| Total aldehydes, ppm | 0.13 | 0.16 | 0.04 | 0.02 | — | 0.07 | 0.08 | 0.08 | 0.07 |
| Formaldehyde, ppm | 0.08 | 0.03 | 0.02 | 0 | — | 0.04 | 0.02 | 0.02 | 0.02 |
| Acrolein, ppm | 0.004 | 0.004 | 0.005 | 0.007 | — | 0.006 | 0.004 | 0.004 | 0.004 |
| PAN, ppm | 0 | 0.05 | 0 | 0 | — | — | — | — | — |
| Eye irr. APCD % | 25.3 | 53.7 | 55.2 | 32.2 | — | — | — | — | — |
| Eye irr. SRI (1) | — | — | — | — | — | — | — | — | 0 |
| Eye irr. SRI (2) | — | — | — | — | — | — | 8.0 | — | — |
| Time: PST 10/4/60 | | | | | | | | | |
| Total aldehydes, ppm | NG | NG | NG | NG | — | 0.14 | 0.12 | 0.08 | 0.05 |
| Formaldehyde, ppm | 0 | 0 | 0 | 0 | — | 0.04 | 0.03 | 0.02 | 0.01 |
| Acrolein, ppm | 0.002 | 0.003 | 0.004 | 0.006 | — | 0.005 | 0.003 | 0.003 | 0.003 |
| PAN, ppm | 0 | 0.04 | 0 | 0 | — | — | — | — | — |
| Eye irr. APCD % | 11.9 | 52.2 | 67.2 | 63.6 | — | — | — | — | — |
| Eye irr. SRI (2) | — | — | — | — | — | — | 5.6 | — | — |
| Time: PST 10/5/60 | | | | | | | | | |
| Total aldehydes, ppm | 0.22 | 0.24 | 0.19 | 0.14 | 0.06 | 0.08 | 0.05 | 0.04 | — |
| Formaldehyde, ppm | 0 | 0.02 | 0 | 0 | 0.01 | 0.02 | 0.01 | 0.01 | — |
| Acrolein, ppm | 0.002 | 0.001 | 0.001 | 0.003 | 0.004 | 0.002 | 0.003 | 0.002 | — |
| Eye irr. SRI (1) | — | — | — | — | — | — | — | — | 0 |
| Eye irr. SRI (2) | — | — | — | — | — | — | 0 | — | — |
| Time: PST 11/1/60 | | | | | | | | | |
| Total aldehydes, ppm | 0.17 | 0.09 | 0.14 | 0.13 | 0.26 | 0.27 | 0.20 | 0.16 | — |
| Formaldehyde, ppm | 0.05 | 0.05 | 0.06 | 0.06 | 0.08 | 0.07 | 0.05 | 0.05 | — |
| Acrolein, ppm | 0.005 | 0.005 | 0.006 | 0.005 | 0.009 | 0.009 | 0.006 | 0.007 | — |
| PAN, ppm | 0 | 0 | 0 | 0 | — | — | — | — | — |
| Eye irr. APCD % | 13.0 | 31.2 | 56.6 | 62.0 | — | — | — | — | — |
| Eye irr. SRI (2) | — | — | — | — | — | — | 20.0 | — | — |
| Time: PST 11/18/60 | | | | | | | | | |
| Total aldehydes, ppm | 0.09 | 0.05 | 0.12 | 0.20 | 0.07 | 0.11 | 0.11 | 0.10 | — |
| Formaldehyde, ppm | 0 | 0 | 0 | 0 | 0.02 | 0.03 | 0.03 | 0.03 | — |
| Acrolein | 0.003 | 0.004 | 0.003 | 0.003 | 0.006 | 0.006 | 0.006 | 0.005 | — |
| Eye irr. SRI (1) | — | — | — | — | — | — | — | — | 2.0 |
| Eye irr. SRI (2) | — | — | — | — | — | — | 8.0 | — | — |

dry gas meter and air pump. The smog sample was drawn through the "sulfur type" absorbers for 10 minutes at 0.75 cubic feet per minute. The analytical procedures for total aldehydes involved taking an aliquot of the above sample and titrating with iodine in one percent starch solution. The formaldehyde analysis was made on the remaining aliquot of the above sample using the chromotropic acid reagent for colorimetric determination at 571 millimicrons.

*2* Sampling procedure and analytical method for acrolein. The sampling apparatus consisted of three "sulfur" absorbers each containing 10 ml of SD-3A alcohol connected in series through vapor traps to a Sprague dry gas meter and air pump. The smog was sampled at 0.1 cubic feet per minute for 30 minutes with the first absorber in an ice water bath and the second and third absorbers in a dry ice-trichloroethylene bath. The analysis was carried out colorimetrically with the use of a reagent (4-hexylresorcinol, mercuric chloride, and trichloracetic acid) and the absorbence measured at 605 millimicrons in 10-cm cells.[7] The District added the reagent to the alcohol in the absorbers before sampling. A sensitivity of one microgram of acrolein was achieved which was approximately three parts per billion in terms of atmospheric concentrations.

*3* The analysis of the 150 liter bag samples for peroxyacylnitrate was based on the infrared spectrum, Fig. 1, of Stephens.[5] The absorbence of 8.6 micron peak was used. The sensitivity of instrument at SRI for these measurements was 0.05 ppm or five percent of full scale on the transmittance chart (400 meter optical absorption path).

*4* The Los Angeles sampling station is also the APCD Smog Station I, where various contaminants are sampled and analyzed continuously. We had available therefore simultaneous observations on the concentration of carbon monoxide by the automatic and continuous nondispersive infrared instrument.

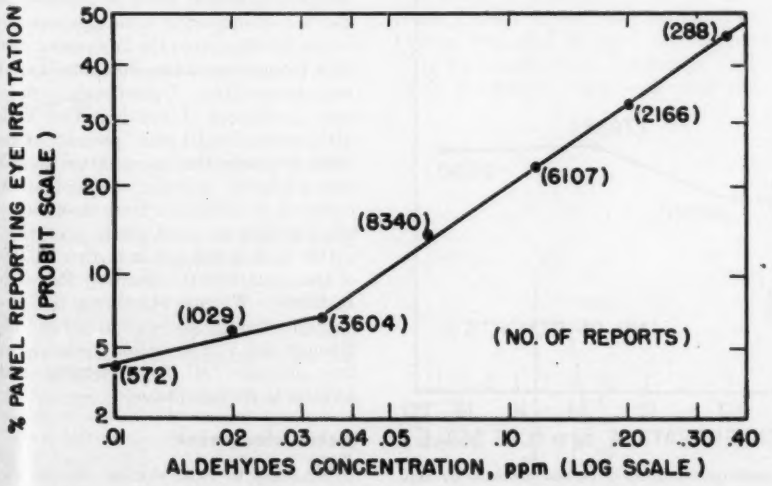


Fig. 2. Regression curve of panel eye irritation on atmospheric concentrations of total aldehydes.

### *Preliminary Results and Discussion*

Table I presents some of the data obtained on this project. These data were selected on the basis that either the total aldehyde concentration exceeded 0.2 ppm (vol) or that eye irrita-

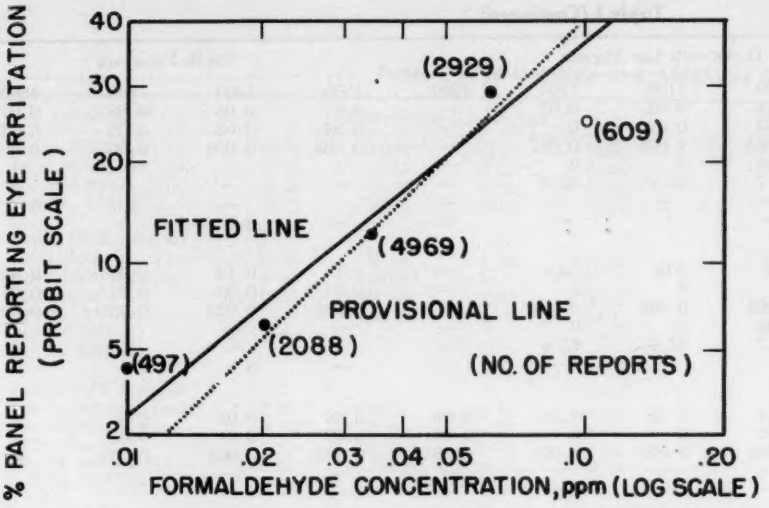


Fig. 3. Regression curve of panel eye irritation on atmospheric concentrations of formaldehyde.

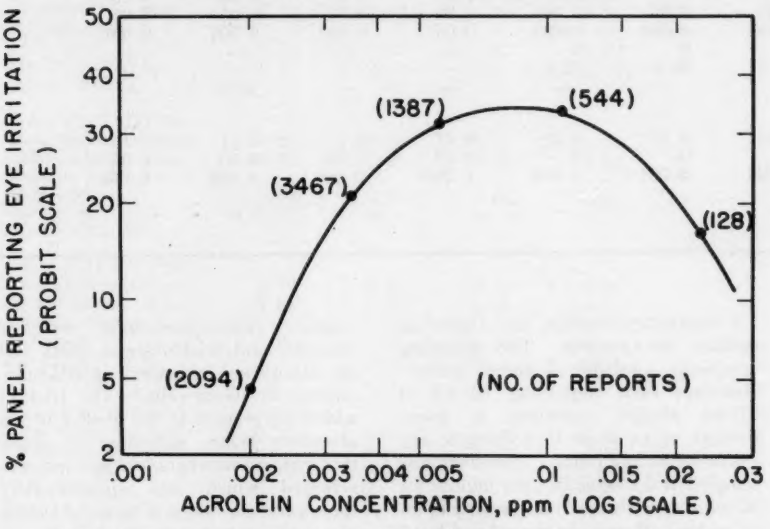


Fig. 4. Regression curve of panel eye irritation on atmospheric concentrations of acrolein.

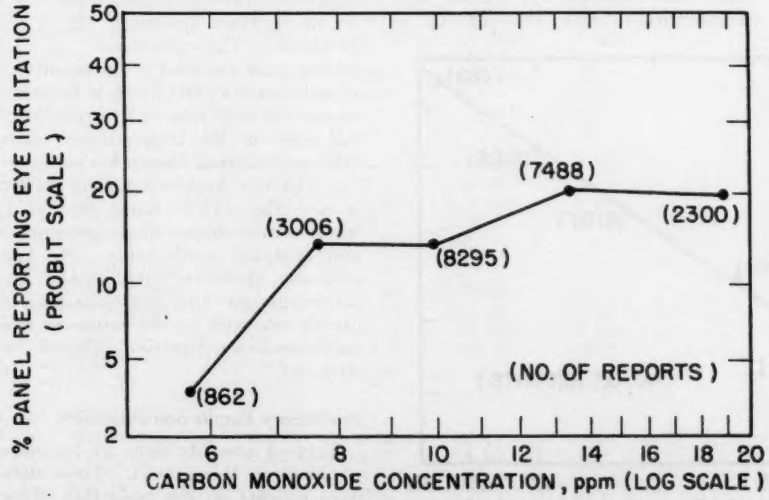


Fig. 5. Regression of panel eye irritation on atmospheric concentrations of carbon monoxide by data groups.

tion index measured in downtown Los Angeles exceeded 50% any time during the daily morning sampling period.

The probit eye irritation is plotted against log concentrations of total aldehydes in Fig. 2. A straight line regression was observed when the data were grouped into five ranges of large sample size.

Figure 3 shows the correlation between formaldehyde concentration and eye irritation although a linear fit in this case was poor compared to that for total aldehydes (provisional line). The line of best fit was calculated by the method of maximum likelihood and the significance of the linear regression relative to deviations from linearity was found to be between 95 and 99% probability.

Figure 4 shows the correlation between acrolein concentration and eye irritation. In this case the relationship is nonlinear although one can draw a smooth curve through the data points.

Figure 5 shows the relationship between carbon monoxide concentration and eye irritation. Since the carbon monoxide level is a measure of the auto exhaust concentration although not necessarily of the irradiation products, it is of interest to note the increase in eye irritation with larger concentrations of carbon monoxide.

Figures 2–5 are plots of all the data taken in downtown Los Angeles from July 5 to Oct. 31, 1960.

The data obtainable for compound X were unfortunately quite meager because the infrared instrument was not available on a regular basis. Furthermore, the concentrations were mostly below 0.05 ppm or just about at or below the lower limit of the analytical method used. It appears that inasmuch as 0.5 ppm is the threshold for eye irritation at 12-minute exposure,[5] this compound was not a serious eye irritant in this atmospheric study. One must qualify this however by stating the formaldehyde and acrolein levels were also low compared to those necessary to produce irritation in the laboratory case even though the atmospheric correlations were respectable. Unfortunately there were incidences of eye irritation with little or no aldehydes present. One must conclude that in addition to the formaldehyde, acrolein, compound X model of eye irritation from smog, some other mechanism must also be operating.

One of us is engaged in further studies of this matter at the Stanford Research Institute. We are examining the hypothesis that photochemical aerosol influences the eye irritation response in the presence of formaldehyde and acrolein in the gas phase.

## Acknowledgment

We wish to acknowledge the assist-

(*Continued on page 427*)

# LIGHT *and* DARK PHASE *Oxidant* PRODUCTION *in a Non-Urban Atmosphere**†

**LYMAN A. RIPPERTON,** Ph.D., Assistant Professor of Sanitary Science, Department of Sanitary Engineering, School of Public Health, University of North Carolina, Chapel Hill, North Carolina

Current meteorological theory[1] postulates that natural‡ ground-level ozone is formed in the upper atmosphere and reaches the ground by diffusion and convection. Artifacticious§ ozone is formed in ground-level air by photochemically initiated oxidation reactions of certain air pollutants.[2] Reasoning by analogy, workers familiar with the chemistry of polluted air have hypothesized that natural ground-level ozone can also be synthesized *in situ* by photochemical reactions.[3]

The purpose of this study was to test the hypothesis that at least part of the natural ground-level oxidant is produced *in situ*.

Chapel Hill, North Carolina, a small non-industrial town in a heavily wooded area, was the site of this study.

In some of the experiments, oxidant production occurred in samples of natural air in the dark with no laboratory irradiation. The dark phase oxidant production generated so much interest and, in some cases, skepticism in our correspondents that a preliminary experimental investigation of the phenomenon has been initiated. Some of the data obtained in that investigation are presented and discussed in this paper.

## Experimental Procedure

### *Natural Air*

Outside air was drawn through a third-story laboratory window into a 72-liter pyrex flask. When flow from the outside was stopped, a Mast ozone meter was attached to the flask in such a manner that the air sample was returned to the flask from the meter (Fig. 1). Some samples were irradiated with a 400-watt mercury vapor lamp (Westinghouse EH-1), but other samples were kept in the dark during the experimental run. (Note: Two thicknesses of pyrex, the glass lamp mantle and the glass flask, absorb those ultraviolet wavelengths which stimulate the production of ozone from diatomic oxygen alone.)

### *Synthetic Atmospheres*

Oxidant concentration was also measured on several synthetic atmospheres during and after irradiation. Several carbonyl compounds were utilized but only data from four runs utilizing isobutyraldehyde at $1^1/_2$ ppm are presented here. The air utilized in the blend was cleansed by being run through activated carbon, drying agent "desiccite," and a cold trap immersed in liquid nitrogen.

Atmospheres containing hexene-1 and $NO_2$ prepared for other studies have also revealed pertinent data. Oxidant concentration in one such system containing two ppm hexene-1 and 1 ppm $NO_2$ was measured at the end of four hours irradiation before and after the light was turned out. Air for this system was cleaned in the same manner as that of the isobutyraldehyde system. The system was static and was blended immediately before irradiation.

A second hexene-1, $NO_2$ system contained one ppm of the hydrocarbon and $^1/_2$ ppm of $NO_2$. Air was cleaned by an activated carbon filter only. This was a continuous flow or dynamic system and the average residence time of the gases in the system was four hours. After a week's operation of the system, the light was turned off and oxidant concentration was measured before and after the system was darkened.

A study[4] was made of the effect of temperature on oxidant production in the irradiated system, air + hexene-1 (2 ppm) + $NO_2$ (1 ppm). Light intensity and pressure were held constant and oxidant concentration was measured at 14, 25, and 35°C.

## Results

### *Natural Air*

Data from the meter's recorder were reduced and concentration values are presented in the accompanying tables as parts per hundred million of ozone. In most instances the value of oxidant in the flask dropped, then rose to a peak, and then subsided.

In nine experiments outside air was

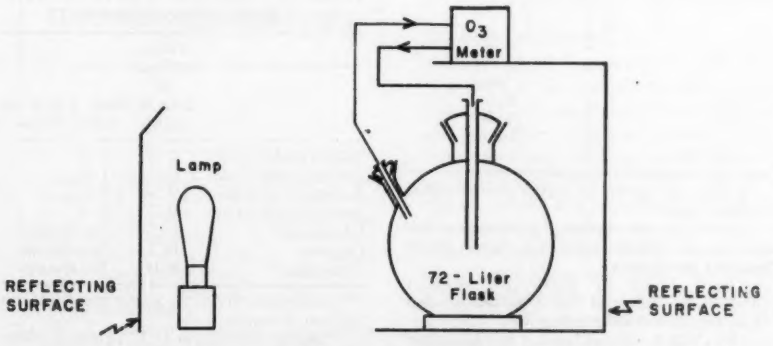


Fig. 1. Irradiation system.

* Presented at the 54th Annual Meeting of APCA, Commodore Hotel, June 11–15, 1961, New York, New York.

† This project was supported in part by the University of North Carolina Research Council and in part by the U. S. Public Health Service, NIH Research Grant No. RG6653(R1).

‡ Natural—Not artificial; in a state of nature, or produced by nature.

§ Artifacticious—Possessing the character of an artifact. (Artifact—a product of artificial nature due to extraneous, especially human, agency.)

**Table I—Oxidant Values in Irradiated Runs**

| Outside Air | Low in Flask Before Irradiation | Peak Value in Flask |
|---|---|---|
| 1.5 | 0.5 | 1.3 |
| 1.0 | 0.3 | 1.8 |
| 0.8 (N) | 0.5 | 1.5 |
| 1.4 | 0.6 | 1.3 |
| 1.4 | 1.0 | 1.4 |
| —[a] | 0.4 | 0.6 |
| 1.0[b] | 0.4 | 0.9 |
| 0.6 (N) | 0.3 | Dropped to 0.0 |
| 0.3 | 0.0 | None |

[a], [b] (N)—See legend, Table II.

irradiated with artificial light after an initial drop of oxidant concentration (Table I). In seven of those nine, the oxidant values rose after irradiation began. In those seven cases, the oxidant value of the outside air ranged from 0.8 to 1.5 parts per hundred million, the low before the peak 0.3 to 1.0, and the peaks from 0.6 to 1.8. The peak in the flask was of the same order of magnitude as the outside value; the ratio of peak value to outside value ranging from 0.87 to 1.98.

Of the eight dark phase runs, five produced peak values of oxidant (Table II). In those five, the air introduced into the flasks contained a range of oxidant values of 0.6 to 1.5, the low in the flask before peaking 0.2 to 1.2, and the peaks 0.5 to 11.0. With one exception, the peak concentration in the flask was of the same order of magnitude as the concentration in the outside air, the ratio (flask peak to outside air) ranging from 0.87 to 2.00. In the exception noted the ratio was 18.33.

Preliminary work with rubber-cracking demonstrated the presence of ozone

**Table II—Oxidant Values in Unirradiated Runs**

| Outside Air | Low in Flask Before Peak | Peak Value in Flask |
|---|---|---|
| 1.5 | 0.3 | 1.3 |
| —[a] | 0.4 | 0.6 |
| 0.4 | 0.5 | 0.8 |
| 1.0[b] | 0.4 | Continuous drop |
| 0.4 | 0.2 | 0.5 |
| 0.6 | 1.2 | 11.0 |
| 0.3 | 0.0 | None |
| 0.0 (N) | 0.0 | None |

Values are given in parts per hundred million ozone.

[a] Dark phase oxidant production followed by irradiation and light phase oxidant production.

[b] Four hours in dark. No oxidant production followed by irradiation and light phase oxidant production.

(N) Night air utilized. In all other runs, daytime air was employed.

in the peak material of the irradiated runs, but did not rule out the possibility that peroxy compounds were also present. Nitrogen dioxide alone in purified air, irradiated or unirradiated, did not produce any of the peaking effects noted. Evidence regarding presence or absence of ozone in the dark peaks was inconclusive.

### *Synthetic Atmospheres*

In the four runs involving isobutyraldehyde, the oxidant value (given in parts per hundred million) (Table III) before and after the end of irradiation were: (light-dark) 1.2–2.0, 0.7–1.9, 0.9–1.1, 0.7–2.6. The clean air alone gave no increase during a two-hour irradiation nor subsequently in the dark for two hours.

When the light was turned out in the static system (hexene-1 at two ppm + $NO_2$ at one ppm) the value of 17 pphm (light phase) began dropping immediately and in 90 minutes reached a value of 7.0 pphm.

When the light was turned off in the dynamic system (hexene-1 at one ppm and $NO_2$ at $^1/_2$ ppm) the oxidant value of 6.4 parts per hundred million (light phase) dropped immediately to 6.2, rose to 6.9, and leveled off at 6.5. After 10 minutes the value began to drop slowly, and at the end of 90 minutes was 5.0 pphm.

The peak concentration of oxidant at one atmosphere and constant light intensity in the system, air + hexene-1 (2 ppm) + $NO_2$ (1 ppm), were: at 14°C 4.4 ppm, at 25°C 11.4 ppm, and at 35°C 29.1 ppm (average of three runs each). (Table IV)

Atmospheres of 100% oxygen and 100% nitrogen had no oxidant increase in two hours irradiation, nor in two hours following irradiation.

## Conclusions and Discussion

The hypothesis that the study originally set out to test was that oxidant material can be synthesized in the lower atmosphere in a non-urban area. The investigator feels that this study has proved the hypothesis in the affirmative.

**Table III—Oxidant Values in Synthetic Atmospheres**

| | Value at End of Irradiation | Peak Value in Dark Phase |
|---|---|---|
| Isobutyraldehyde[a] | 1.2 | 2.0 |
| Isobutyraldehyde | 0.7 | 1.9 |
| Isobutyraldehyde | 0.9 | 1.1 |
| Isobutyraldehyde | 0.7 | 2.6 |
| Clean air | 0.7 | No change |
| Oxygen | 0.3 | No change |
| Nitrogen | 0.0 | No change |

Values are given in parts per hundred million of ozone.

[a] Isobutyraldehyde at $1^1/_2$ ppm in clean air.

**Table IV—Peak Oxidant Values at Various Temperatures in the System, Hexene-1 + $NO_2$ at Constant Pressure and Light Intensity**

| Temperature, °C | Experimental Run 1 | 2 | 3 |
|---|---|---|---|
| 14 | 4.0 | 6.2 | 2.9 |
| 25 | 10.0 | 12.2 | 12.1 |
| 35 | 29.2 | 29.4 | 28.7 |

Values are given in parts per hundred million of ozone.

In testing the hypothesis, the phenomenon of dark phase oxidant synthesis was "rediscovered." (The term "rediscovered" is used advisedly because substantial, if not conclusive, proof of the dark phase synthesis existed prior to 1860.[5])

Nitrogen dioxide plus air-borne organic compounds, including hydrocarbons and aldehydes, from forest trees and other plants could be the oxidant precursors. Irradiated aldehydes alone will produce oxidant material. One is tempted to attribute oxidant production to photochemical reactions of the same type as those occurring in the polluted atmosphere of Los Angeles. Went has theorized that ozone and oxidant in the lower atmosphere are products of the photochemical oxidation of organic compounds, particularly terpenes from plants.[3] The dark phase reactions observed, however, prevent the complete equation of these reactions with those of the Los Angeles atmosphere. No such dark reactions have been reported there, but this dissimilarity may be a function of either relative or absolute concentration.

### *Purpose of Reporting Results*

The purpose of reporting some of the results of dark phase oxidant production in synthetic atmospheres at this time was principally to demonstrate that something similar to the natural phenomenon can be reproduced in the laboratory. The fact that some steps in the so-called "photochemical" synthesis of oxidant are obviously "thermal" offers a possible explanation for the dark phase oxidant synthesis observed. The series of reactions leading to oxidant production can be initiated by a photochemical effect. Some of the subsequent steps, however, apparently are not light-dependent and can proceed in the dark.

The presence of aldehydes and their concentration, relative and absolute, may be an important factor in the dark oxidant production, as demonstrated by the fact that irradiation of aldehydes before they were placed in the dark resulted in oxidant production in the dark conditions. In the static system of hexene-1 and $NO_2$ oxidant concentration drops immediately after the light is removed; this reaction has been

going on for four hours. In the dynamic system of hexene-1 and $NO_2$ oxidant concentration was maintained for 10 minutes before dropping, indicating a continued oxidant production during that time. In this latter case the average residence time of reactants is four hours, but some of the effluent has been in the system for much longer. Assuming complete instantaneous mixing, at least 36% of the material leaving the dynamic system will have had a residence time greater than four hours, according to the first order equation, so that a system such as this would change chemically with age. The difference in behavior between these two hydrocarbon, $NO_2$ systems first suggested to the investigator that the accumulation of some reaction products, like carbonyl compounds, might be important in the dark phase reaction. The exact role of carbonyl compounds in the dark phase oxidant production must await further elucidation, but they seem to be, if not intermediates, closer to the immediate precursors than hexene-1 and $NO_2$.

The fact that there is continuation of oxidant production for a while after the removal of light caused by a series of "thermal" reactions explains some but not all of the dark phase phenomena observed. Further work is contemplated.


### Summary

Ozone and oxidant production was demonstrated to take place in natural ground-level air which had been drawn into a 72-liter flask. This production was sufficient to produce a concentration of oxidant of the same order of magnitude as that found in the ambient outdoor air.

Oxidant production occurred both in natural air which was irradiated with visible light in the laboratory and in that which was not irradiated.

A similar dark phase oxidant production was observed in synthetic atmospheres. In these cases the reactants were first irradiated in the laboratory and oxidant production was observed in the subsequent dark phase. The use of carbonyl compounds in these experiments holds promise, and one of the synthetic atmospheres utilized in this study which showed dark phase oxidant production was clean air $+1^1/_2$ ppm of isobutyraldehyde.


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## ATMOSPHERIC SAMPLING

(*Continued from page 424*)

ance of James Orcutt and James R. Taylor of APCD, in the statistical analyses; and E. A. Schuck and N. Endow of Stanford Research Institute and H. Mayrsohn of the APCD, in the analytical phases of the project.

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**COMPLETE SETS OF ABSTRACTS and 1960 PROCEEDINGS AVAILABLE THROUGH HEADQUARTERS**


## MATTER RELATIONSHIP

(*Continued from page 407*)

nonurban pollution is, in reality, merely highly diluted urban pollution.


### Summary

The relationships between benzene soluble organic matter and suspended particulate matter in air particulate samples collected at 144 urban and 38 nonurban stations of the National Air Sampling Network, during 1957 and 1958, have been compared, using arithmetic means, geometric means, and standard geometric deviations as a basis for the comparison.

The national arithmetic mean value of the benzene soluble organic to suspended particulate ratio was 0.082 for the urban stations and 0.084 for the nonurban stations, and the respective geometric mean values were 0.074 and 0.066. Standard geometric deviations of 1.62 for urban, and 1.98 for nonurban stations, indicate a greater range of ratio values for nonurban stations.

Note: A detailed tabulation of all data used in the preparation of this paper may be obtained from the authors upon request.


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## PARTICULATE ANALYSIS

(*Continued from page 409*)

on the Research Appliance smoke sampler and the Staplex high volume sampler operating for one hour. Successful determinations have been made on membrane filters and filter paper from the M.S.A. Samplair when loadings have been visibly high. The ash from soot-fall samples has also been subjected to this procedure with a high degree of accuracy. If necessary analysis for Ca, Al, Fe, Si, Mg, and Ge can be included.

#### *Accuracy of Method*

The method is generally accurate from 10 to 20% of the component determined. Usually this is resolved when a different spectrum line of Indium is used in the intensity ratio. A large source of error is due to currents of air in the immediate vicinity of the arc stand while the firing procedure is in progress. For best results this operation should be conducted in a draft free and dust free room. With considerable care and attention to details, and after experience with the procedure, an accuracy of 10% or better is possible.

# A RELIABLE LOW COST INSTRUMENT *for Determining Atmospheric Oxidant Levels**

**ROSS V. DOUGHTY,** Engineer, **DAVID O. ERISMAN,** Chemist, Bureau of Combustion Control, City of Dayton, Dayton, Ohio

In recent years, community air monitoring groups have shown increasing interest in the quantitative measurement of atmospheric ozone or oxidants. The presence of ozone as an air pollutant is generally attributed to a photochemical reaction involving sunlight, hydrocarbons, and oxides of nitrogen. Considerable data have been collected indicating a definite relationship between high ozone concentrations and smog severity in the Los Angeles area. Although ozone is by no means the only oxidant occurring as an air pollutant, it is by far the most common. Many areas having serious air pollution problems do not experience high oxidant concentrations since they have predominantly reducing atmospheres. The preponderance of reducing agents, such as sulfur dioxide, in such areas, reacts with the oxidizing agents, resulting in little or no remaining atmospheric oxidant. It is the belief of the authors that, in the future, many areas will experience apparent increased oxidant levels, as reducing agent levels are lowered.

The community contemplating a survey of oxidant levels has a wide variety of methods from which to choose. However, in many instances, particularly in smaller communities, the choice of a suitable method is governed mainly by the cost involved.

Automatic instrumental methods providing continuous sampling and recording are beyond the budget capabilities of many communities. Wet chemical methods, while widely used, require laboratory facilities and trained personnel which are not always available. In addition, extensive field sampling is often difficult with wet methods due to unstable reagents, lack of power facilities at sampling sites, and the necessity of returning samples to the laboratory for analysis. These limitations can be quite frustrating to the control agency operating on a low budget.

Many communities utilize rubber cracking methods such as that of Crabtree and Kemp,[1] or Bradley and Haagen-Smit.[2] These methods have advantages such as simplicity and low cost. However, the results obtained may vary considerably depending upon the experience and interpretative abilities of the operator. Since the visual observation of cracking is generally taken as the end point of the test, the personal judgment of the operator becomes a variable of major importance.

Of the common methods, it would seem that rubber cracking would hold the most promise for the smaller community agency, hampered by limited personnel and facilities. This paper is concerned with the improvement of the rubber cracking method by removal of the element of varying individual judgment, thereby increasing the accuracy and reliability of the method and permitting a more valid comparison of the

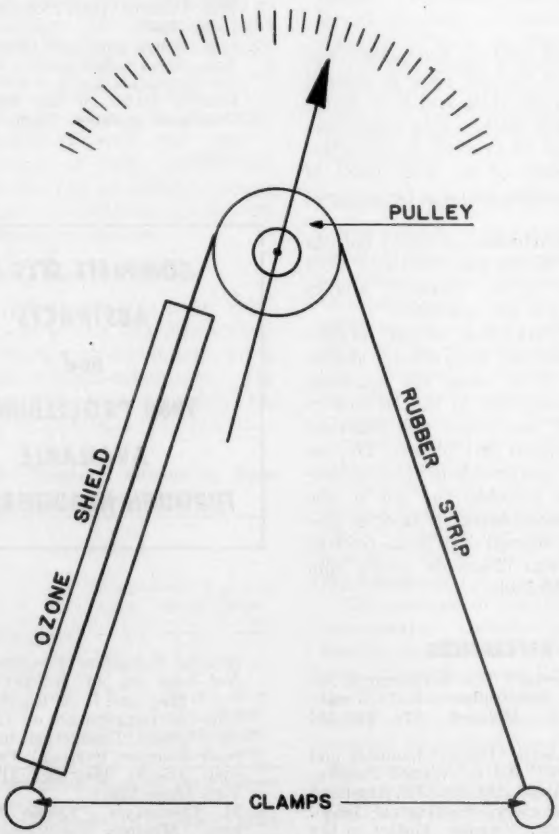


Fig. 1.

* Presented at the 54th Annual Meeting of APCA, Commodore Hotel, June 11–15, 1961, New York, New York.

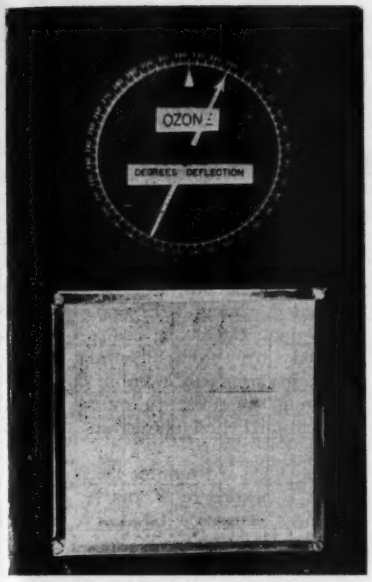


Fig. 2. Ozone detection instrument, front view.

data produced by different operators.

### Instrument Design

Several authors have proposed methods for measuring oxidant induced rubber cracking as a function of creep in a strip of rubber under stress.[3, 4] After investigating various techniques utilizing this concept, a method similar to that used by the B. F. Goodrich Ozonometer was selected as the basis for this study.[4] This method measures the differential creep in a standard strip of rubber, one half of which is exposed to the atmosphere and the other half of which is protected. Constant stress is maintained in both halves of the rubber strip by a pulley located at the median point of the strip. Oxidant induced creep is measured by the degree of rotation of the pulley. A schematic diagram of the instrument is shown in Fig. 1. The ozone shield is lined with rubber or cellulose to bring about the rapid decomposition of ozone and thus protect the enclosed standard rubber strip. The elongation of the rubber strip is approximately 100%.

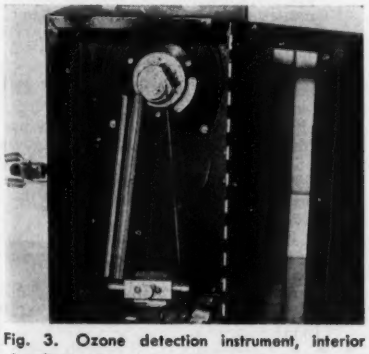

Fig. 3. Ozone detection instrument, interior view from rear.

In test use, the ends of the standard rubber strip are fastened in the clamps and the strip stretched over the pulley. The ozone shield is closed and the instrument is exposed to the atmosphere to be sampled. In approximately five minutes the instrument will reach equilibrium with the air temperature and the indicator can be set at zero and the time noted. Indicator deflection is recorded at convenient time intervals throughout the sampling period. Ozone concentration is determined from the calibration curve by plotting deflection versus time.

### Development

Initial exploratory work was conducted with a prototype instrument. This instrument, it was found, did not lend itself too well to the rigors of field use. Sensitivity was limited and reproducible results could not always be obtained. Accordingly, with the experience thus gained, a new instrument was built, modifying the original design to provide increased ruggedness, sensitivity, and reliability. The relatively heavy indicating pointer of the original design was replaced with a light weight, well balanced needle mounted in ball bearings and fitted with a readily adjustable degree plate. The unit was enclosed in a ventilated metal case to provide protection from the weather. The improved instrument proved much less sensitive to position and external shock which made its use more foolproof in the field. The response of the instrument to low concentrations of ozone was also very much increased. The improved instrument is shown in Figs. 2 and 3.

### Calibration Procedures

Calibration of the instrument was carried out by subjecting it to known concentrations of ozone in a test chamber. The chamber used had a volume of approximately 1.5 cu ft and was constructed of aluminum. One side of the chamber was provided with a glass window for observation of the interior. Prior to use, the chamber was "conditioned" with heavy doses of ozone from an electrical discharge type ozonator to minimize ozone reaction with the chamber itself during later tests. Ozone for test use was furnished by a generator consisting of four General Electric G-4S-11 ultraviolet ozone lamps enclosed in a glass container. The ozone output of the generator was controlled by the number of lamps in operation. A measured volume of air was passed through the generator and into the chamber. Constant air flow was maintained by a vacuum pump connected to the chamber exhaust. A small fan in the chamber provided thorough mixing of the test atmosphere.

Several methods were used at different times to monitor the ozone concentration within the chamber. The most consistent results were obtained with a Mast oxidant recorder,[5] sampling through a probe located in the center of the chamber. Chamber samples were also bubbled through potassium iodide solution and titrated with 0.001 *N* sodium thiosulfate to determine ozone.[6] Attempts were made to utilize the phenolphthalin method of Haagen-Smit for chamber monitoring, although there is some disagreement as to the reliability of this method, and trustworthy results were seldom obtained.[7] When sufficient data had been accumulated on the behavior of the rubber cracking instrument in the test chamber a calibration graph was prepared, plotting chamber concentration versus degrees deflection per minute as indicated by the instrument. Standard

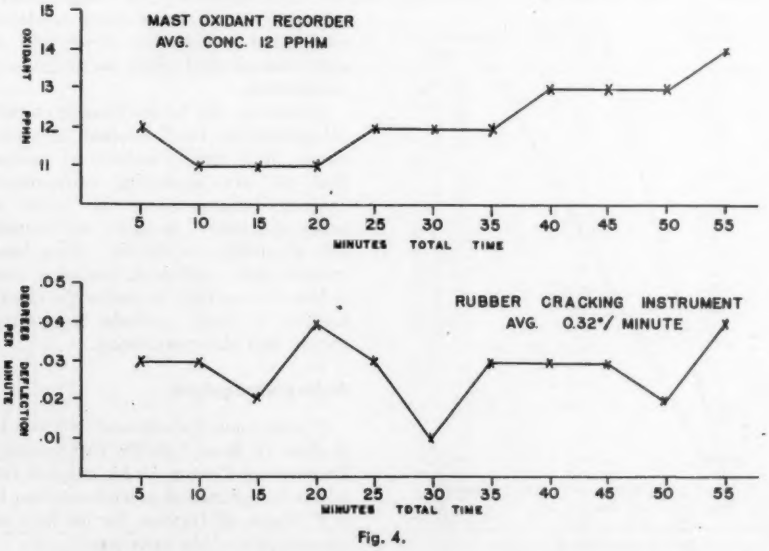


Fig. 4.

rubber strips for use in the instrument were obtained from the B. F. Goodrich Company.[8] Any variation in the composition, or cross sectional area of the rubber strips used would require recalibration of the instrument.

It was found during calibration that air temperature affected the response of the instrument. A series of calibration runs was made at varying chamber temperatures and results plotted for each temperature. Atmospheric temperature must be taken into account when determining cumulative ozone levels from the calibration curve. Normal variations in relative humidity did not appreciably affect instrument response, although slightly increased deflection was noted with very high humidity.

Results of a typical calibration run at 80°F are shown in Fig. 4 where the Mast recorder indicates an average chamber concentration of 12 pphm versus an average of 0.32° deflection per minute for the rubber cracking instrument. A completed calibration curve for two different temperatures is shown in Fig. 5.

Daily atmospheric measurements have been taken for over a year with the rubber cracking instrument in Dayton, Ohio. Monthly cumulative oxidant levels as measured are listed in Table I.

In chamber tests at relatively high concentrations, it was found that total deflections of approximately 250° could be obtained before excessive cracking caused the rubber strip to break. This situation would never be encountered at normal atmospheric levels using a reasonable sampling period. Furthermore, it is unlikely that the accuracy of the method could be maintained at such extreme deflections, although further investigation is needed in this area. It was also found that measurements could not be made at temperatures below 35 to 40°F with any degree of confidence. At low temperatures, rubber under stress tends to crystallize and lose its susceptibility to ozone.

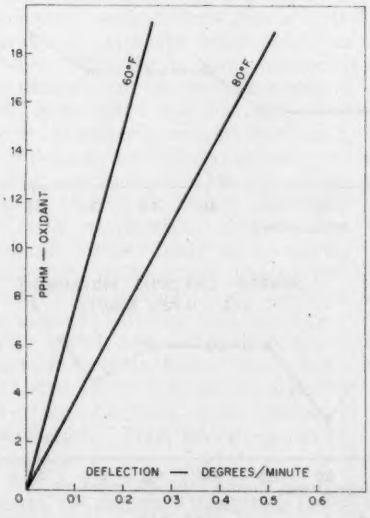

Fig. 5.

**Table I—PPHM Oxidant, Dayton, Ohio —1960**

| Month | Range | Average |
|---|---|---|
| January | 0.1–2.3 | 0.3 |
| February | 0.1–1.5 | 0.3 |
| March | 0.1–1.8 | 0.9 |
| April | 0.5–2.2 | 1.2 |
| May | 0.1–1.8 | 0.9 |
| June | 0.3–1.9 | 1.0 |
| July | 0.4–1.8 | 1.2 |
| August | 0.6–1.7 | 1.1 |
| September | 0.2–1.5 | 0.9 |
| October | 0.3–1.0 | 0.6 |
| November | 0.2–0.7 | 0.5 |
| December | 0.2–0.5 | 0.3 |

## Conclusion

It is firmly believed that the instrument described in this paper can provide accurate, low-cost measurement of cumulative oxidant levels. The simple, virtually foolproof operation of the instrument eliminates the need for trained personnel and frequent maintenance. The instrument requires no power source and thus is readily applied to field sampling at isolated sites.

In final evaluation, it must be remembered that all of the common methods for the determination of atmospheric oxidants are empirical. All of these methods depend upon a measurement in terms of the oxidation of a reference substance such as potassium iodide or rubber, rather than a direct measurement of oxidants. Thus, because the reference substances themselves are different, we cannot hope to obtain numerically identical results with different methods. We must choose one method with one reference substance and use it consistently if we wish to obtain meaningful results for purposes of comparison.

Moreover, due to the present empirical nature of most oxidant measurements, it is hardly correct to assume that we are measuring atmospheric oxidant concentrations in terms of actual amounts. In effect, we measure only a condition of the air. This, however, is quite sufficient, assuming consistency of method, to enable the identification of peak periods, long-range trends, and other variations.

## Acknowledgment

Grateful acknowledgment is given to Andrew H. Rose, USPHS Taft Sanitary Engineering Center, for his support and advice in calibration procedures, and to J. P. Owen, of Dayton, for his help on construction of the instrument.

## PHS ESTABLISHES NEW OFFICE AS STEP IN REORGANIZATION

The establishment of an Office of Resource Development (Environmental Health) within the Bureau of State Services was announced recently, as another step in the reorganization of the Public Health Service to provide, among other purposes, a more coordinated, integrated, and intensive focus upon the public health problems created by environmental hazards stemming primarily from air and water pollution, exposure to radiation, and occupational and industrial developments.

The Office of Resource Development, which is headed by Frank A. Butrico, Engineer, Director Grade, is a staff arm to Dr. Robert J. Anderson, Assistant Surgeon General and Deputy Chief of the Bureau of State Services.

### Training Branch Established

In an effort to stimulate greater research efforts by the life and engineering sciences in the many aspects of environmental health hazards, a research and/or training grants branch has been established in a number of Divisions within the Bureau, as follows: Division of Air Pollution, Chief, Research and Training Grants Branch, Dr. Maurice Bender; Division of Environmental Engineering and Food Protection, Chief, Research Grants Branch, Harold B. Robinson; Division of Occupational Health, Acting Research Grants Administrator, Dr. W. Clark Cooper; Division of Radiological Health, Chief, Office of Extramural Grants, Dr. Paul F. Hahn; and Division of Water Supply and Pollution Control, Chief, Research and Training Grants Branch, Harry A. Faber.

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# The BEHAVIOR of Dense STACK GASES*

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Due to the need in Du Pont to provide satisfactory air pollution abatement for the discharge of heavier-than-air gases, the behavior of such emissions in the atmosphere has been investigated. Atmospheric dispersion equations and techniques have been available[1-10] to calculate ground level concentrations of contaminants from stack discharges that have the same or less density than ambient air. Chesler and Jesser[11] and the writer[5] have observed and discussed the descent of dense stack gases. Scorer[12] has discussed plume descent from evaporative cooling of entrained water in stack gases. Also, Bosanquet[13] has proposed a formula for the net rise of a dense plume. Additional knowledge was required, however, to enable assessment of the odor, toxicity, and explosion potential of discharges of dense gases at points downwind from the source.

In the planning for the proposed study, large-scale field tests were considered but it was concluded that they would be too difficult and excessively time-consuming. Therefore, model tests on the behavior of dense stack gases were performed. The tests were carried out under contract by the Du Pont Company with Research Division of New York University, and were performed by their personnel in the $3^1/_2$ feet high by seven feet wide wind tunnel. Du Pont personnel prepared the specifications of test conditions and analyzed the data.

## Description of Model Tests

Strom and his NYU co-workers[14-16] have discussed the application of model tests to air pollution studies. For geometrical similarity between the stack gases in the tunnel and full scale, the scale factor for velocity, $V$; length, $L$; and acceleration due to gravity, $g$, are related to the equality of Froude's number, $V^2/Lg$, in the tunnel and full scale. In the present series of tests, the linear scale was 4 ft = 1 in., or

$$\frac{L_{\text{tunnel}}}{L_{\text{full scale}}} = \frac{1}{48} = 0.021$$

$$\frac{V_{\text{tunnel}}}{V_{\text{full scale}}} = \frac{1}{(48)^{0.5}} = 0.145$$

$$\frac{Q_{\text{tunnel}}}{Q_{\text{full scale}}} = \left(\frac{L_{\text{tunnel}}}{L_{\text{full scale}}}\right)^{2.5} = 6.2 \times 10^{-5}$$

where $Q$ = flow rate (volume/time).

The model tests are representative of a neutral (adiabatic) temperature gradient in a full scale atmosphere, since no thermal modification of the wind tunnel air stream was employed.

The following specific gravities were selected for study: 1 (air), 1.17, 1.52, 2.96, and 5.17.

Stack gas composition was achieved by proper mixture of "Freon-114,"† which has a specific gravity of 5.9, and air.

"Freon-114"† is nontoxic and nonflammable and was an ideal gas for efficient and safe performance of the tests. It is important to emphasize that the indicated specific gravities refer to the total gas stream, not to a high molecular weight constituent, which in many cases will exist in stack gases in diluted form.

Combinations of full-scale stack diameters, stack gas velocities, and wind speeds used in the tests are listed in Table I. Use of a constant stack height scaled for 100 feet was attempted. In some tests, as noted in Table I, stack height had to be reduced to 60 feet, since with a 100 feet stack the plume hit the tunnel ceiling at high stack gas velocities. Three miles per hour was the lowest wind speed that could be tested.

The stack effluents were made visible by an oil-fog "smoke." For evaluation of the tests, the visible stack plumes were photographed at selected intervals so that the results represent average conditions during approximately one minute.

## Results of Tests

Graphs of the center-line of the plumes were prepared from the photographic negatives. In all cases, the dense stack gases descended to the ground. Data in Table II show the distance from the stack where the lower

**Table I—Combination of Test Variables (Full-Scale)**

| Molecular Weight | Specific Gravity, Air = 1 | Stack Gas Velocity, Ft/Sec | Wind Speed, Mph | Stack Diameter, Ft | Stack Height, Ft |
|---|---|---|---|---|---|
| 29 | 1 | 20 | 3,6,10 | 1 and 2 | 100 |
| 29 | 1 | 50 | 3,6,10 | 1 | 100 |
| 29 | 1 | 50 | 10 | 2 | 100 |
| 29 | 1 | 50 | *,6,10 | 2 | 60 |
| 29 | 1 | 80 | 3,6 | 1 | 60 |
| 29 | 1 | 80 | 10 | 1 | 100 |
| 29 | 1 | 80 | *,6,10 | 2 | 60 |
| 34 | 1.17 | 20 | 3,6,10 | 1 and 2 | 100 |
| 34 | 1.17 | 50 | 3,6,10 | 1 and 2 | 100 |
| 34 | 1.17 | 100 | 3,6,10 | 1 | 60 |
| 34 | 1.17 | 100 | *,6,10 | 2 | 60 |
| 44 | 1.52 | 20 | 3,6,10 | 1 and 2 | 100 |
| 44 | 1.52 | 50 | 3,6,10 | 1 and 2 | 100 |
| 44 | 1.52 | 80 | 3,6,10 | 1 | 100 |
| 44 | 1.52 | 80 | 6,10 | 2 | 100 |
| 44 | 1.52 | 80 | 3,6,10 | 2 | 60 |
| 86 | 2.96 | 20 | 3,6,10 | 1 and 2 | 100 |
| 86 | 2.96 | 50 | 3,6,10 | 1 and 2 | 100 |
| 86 | 2.96 | 150 | 3,6,10 | 1 | 60 |
| 86 | 2.96 | 100 | 3,6,10 | 2 | 60 |
| 150 | 5.17 | 20 | 3,6,10 | 1 and 2 | 100 |
| 150 | 5.17 | 50 | 3,6,10 | 1 and 2 | 100 |
| 150 | 5.17 | 80 | 3,6,10 | 1 | 100 |
| 150 | 5.17 | 80 | 3,6,10 | 2 | 60 |

* Scheduled, but not performed since plume hit tunnel ceiling for 60 ft stack height.

* Presented at the 54th Annual Meeting of APCA, Commodore Hotel, June 11-15, 1961, New York, New York.

† Du Pont registered trademark for fluorinated hydrocarbon refrigerants.

edge of the plume struck the ground.

Photographs for selected combinations of test variables are shown in Figs. 1 and 2. The important difference in these figures is stack gas velocities of 20 and 80 ft/sec in Fig. 1 and Fig. 2, respectively. Even with a stack gas velocity as high as 80 ft/sec, however, there is rapid plume descent for the indicated specific gravities of 1.52 and 5.17.

*Initial Plume Rise.* The following expression was developed for the initial plume rise of the dense stack gas:

$$h_r = 5.44\ (D)^{0.5} \left(\frac{V_s}{u}\right)^{0.75} \qquad (1)$$

where:

$h_r$ = initial rise of plume, ft
$D$ = stack diameter, ft
$V_s$ = stack gas velocity, ft/sec
$u$ = horizontal wind speed, ft/sec

The above formula was verified for goodness of fit by the chi-square test.[17] Equation (1) is statistically significant for specific gravities 2.96 and 5.17 but not for specific gravities 1.17 and 1.52 for a stack diameter of one foot. Equation (1) is highly reliable for the plume center-line for a two foot diameter stack, if the four tests with a combination of three mph wind speed and 20 ft/sec stack gas velocity are omitted. The expression of Equation (1) gives initial rises that are much too large for combinations of three mph wind speed and 20 ft/sec stack gas velocity. This low wind speed—low stack gas velocity plume rise exclusion—is of little practical significance, since in the Conclusion section to this paper, it is stated that it is best practice to eject dense stack gases at high velocities.

A major significant result is that the initial rise of dense plumes is less than for air, for which model tests were also performed, even though the momentum of the dense plumes is greater than air. This behavior is apparently the result of decelerating buoyant forces in the initial ascent. Also, the initial rise of dense stack gases is virtually independent of specific gravity. This indicates that the rate of dissipation of momentum is proportional to the decelerating forces of buoyancy.

*Rates of Plume Descent.* The acceleration of the plume due to buoyancy is:

$$a = g\left(\frac{1 - \text{Sp. Gr.}}{\text{Sp. Gr.}}\right) \qquad (2)$$

where:

$a$ = acceleration, ft/sec$^2$
$g$ = acceleration due to gravity, 32.2 ft/sec$^2$
Sp. Gr. = specific gravity of the plume where air = 1

The downward acceleration of the plume at the terminal point of the initial

**Table II—Distance from Stack Where Lower Edge of Plume Touches Ground**

| Specific Gravity, Air = 1 | Stack Gas Velocity, Ft/Sec | Wind Speed, Mph | Stack Diameter, Ft | Stack Height, Ft | Distance Where Lower Edge of Plume Touches Ground, Ft |
|---|---|---|---|---|---|
| 1.17 | 20 | 3 | 1 | 100 | 230 |
| 1.17 | 20 | 6 | 1 | 100 | 790 |
| 1.17 | 20 | 10 | 1 | 100 | 650 |
| 1.17 | 50 | 3 | 1 | 100 | 330 |
| 1.17 | 50 | 6 | 1 | 100 | 780 |
| 1.17 | 50 | 10 | 1 | 100 | 830 |
| 1.17 | 100 | 3 | 1 | 60 | 400 |
| 1.17 | 100 | 6 | 1 | 60 | 820 |
| 1.17 | 100 | 10 | 1 | 60 | 810 |
| 1.17 | 20 | 3 | 2 | 100 | 190 |
| 1.17 | 20 | 6 | 2 | 100 | 830 |
| 1.17 | 20 | 10 | 2 | 100 | 800 |
| 1.17 | 50 | 3 | 2 | 100 | 190 |
| 1.17 | 50 | 6 | 2 | 100 | 820 |
| 1.17 | 50 | 10 | 2 | 100 | 820 |
| 1.17 | 100 | 6 | 2 | 60 | 820 |
| 1.17 | 100 | 10 | 2 | 60 | 820 |
| 1.52 | 20 | 3 | 1 | 100 | 190 |
| 1.52 | 20 | 6 | 1 | 100 | 800 |
| 1.52 | 20 | 10 | 1 | 100 | 800 |
| 1.52 | 50 | 3 | 1 | 100 | 210 |
| 1.52 | 50 | 6 | 1 | 100 | 810 |
| 1.52 | 50 | 10 | 1 | 100 | 810 |
| 1.52 | 80 | 3 | 1 | 100 | 400 |
| 1.52 | 80 | 6 | 1 | 100 | 800 |
| 1.52 | 80 | 10 | 1 | 100 | 815 |
| 1.52 | 20 | 3 | 2 | 100 | 130 |
| 1.52 | 20 | 6 | 2 | 100 | 440 |
| 1.52 | 20 | 10 | 2 | 100 | 850 |
| 1.52 | 50 | 3 | 2 | 100 | 200 |
| 1.52 | 50 | 6 | 2 | 100 | 650 |
| 1.52 | 50 | 10 | 2 | 100 | 820 |
| 1.52 | 80 | 3 | 2 | 60 | 175 |
| 1.52 | 80 | 6 | 2 | 60 | 440 |
| 1.52 | 80 | 10 | 2 | 60 | 620 |
| 1.52 | 80 | 6 | 2 | 100 | 700 |
| 1.52 | 80 | 10 | 2 | 100 | 800 |
| 2.96 | 20 | 3 | 1 | 100 | 165 |
| 2.96 | 20 | 6 | 1 | 100 | 550 |
| 2.96 | 20 | 10 | 1 | 100 | 760 |
| 2.96 | 50 | 3 | 1 | 100 | 150 |
| 2.96 | 50 | 6 | 1 | 100 | 595 |
| 2.96 | 50 | 10 | 1 | 100 | 800 |
| 2.96 | 150 | 3 | 1 | 60 | 200 |
| 2.96 | 150 | 6 | 1 | 60 | 410 |
| 2.96 | 150 | 10 | 1 | 60 | 800 |
| 2.96 | 20 | 3 | 2 | 100 | 100 |
| 2.96 | 20 | 6 | 2 | 100 | 210 |
| 2.96 | 20 | 10 | 2 | 100 | 460 |
| 2.96 | 50 | 3 | 2 | 100 | 140 |
| 2.96 | 50 | 6 | 2 | 100 | 350 |
| 2.96 | 50 | 10 | 2 | 100 | 500 |
| 2.96 | 100 | 3 | 2 | 60 | 150 |
| 2.96 | 100 | 6 | 2 | 60 | 250 |
| 2.96 | 100 | 10 | 2 | 60 | 540 |
| 5.17 | 20 | 3 | 1 | 100 | 110 |
| 5.17 | 20 | 6 | 1 | 100 | 370 |
| 5.17 | 20 | 10 | 1 | 100 | 670 |
| 5.17 | 50 | 3 | 1 | 100 | 120 |
| 5.17 | 50 | 6 | 1 | 100 | 800 |
| 5.17 | 50 | 10 | 1 | 100 | 800 |
| 5.17 | 80 | 3 | 1 | 100 | 210 |
| 5.17 | 80 | 6 | 1 | 100 | 450 |
| 5.17 | 80 | 10 | 1 | 100 | 800 |
| 5.17 | 20 | 3 | 2 | 100 | 70 |
| 5.17 | 20 | 6 | 2 | 100 | 230 |
| 5.17 | 20 | 10 | 2 | 100 | 400 |
| 5.17 | 50 | 3 | 2 | 100 | 70 |
| 5.17 | 50 | 6 | 2 | 100 | 265 |
| 5.17 | 50 | 10 | 2 | 100 | 630 |
| 5.17 | 80 | 3 | 2 | 60 | 80 |
| 5.17 | 80 | 6 | 2 | 60 | 170 |
| 5.17 | 80 | 10 | 2 | 60 | 370 |
| 5.17 | 80 | 10 | 2 | 100 | 480 |

ascent is due to negative buoyancy. Equation (2) indicates that small forces can initiate downward motion. For example, a specific gravity of the plume of only 1.002 at the terminal point of the initial ascent will result in an acceleration of −0.064 ft/sec.[2] This acceleration acting solely through a 100 ft vertical distance would result in a final descending velocity of 3.6 ft/sec. The additional complex forces that result from interaction of the plume and ambient air would decrease the magnitude of the acceleration. The rates of plume descent from the model tests, as discussed below, are of most use and interest, however. These rates can be determined from the graphs of the effluent center-line. Figs. 3 to 5 show velocities of plume descent for a two foot diameter stack for 20, 50, and 80–100 ft/sec stack gas exit velocities. The descending velocities in these figures, which are one minute averages, are for neutral stability at the height of maximum descending speed. Velocities of plume descent increase generally with an increase in specific gravity of the stack gas. Seven to eight ft/sec is the limiting maximum velocity of descent with a two foot diameter stack for the range of specific gravities that was evaluated. For specific gravity equal to or greater than two, the rates of descent are 1.5 to two times greater for a two foot diameter stack than for a one foot diameter stack. This is because of less dilution and greater negative buoyancy for the two foot diameter stack.

*Dilutions at the Ground.* Measurements of the number of dilutions or actual concentrations reaching ground-level was beyond the scope of the present study. An estimate of dilutions of the stack effluent at the ground can be made by assuming the cross-sectional area of the plume to be circular. By comparing the cross-sectional area of the stack and the plume where the lower edge of the latter strikes the ground, dilutions there can be estimated: Dilution = (plume diameter/stack diameter).[2] Three miles per hour gave the least dilution of the wind speeds, three, six, 10 mph, that were tested. Figs. 6 and 7 show dilutions of the stack gas at the ground for stack diameters of one foot and two feet respectively, for three mph wind speed, neutral stability, and specified stack gas variables. The dilutions are averages over one minute and across the diameter of the plume. The least dilutions in the center of the plume would be approximately one-half of the values given in Figs. 6 and 7 on the basis of wind tunnel studies by others.[18]

An examination of Figs. 6 and 7 indicates that other things being equal, the least dilution at the ground is approximately inversely proportional to the cross-sectional area, or square of the diameter of the stack, and accordingly, to the effluent discharge rate. This relationship indicates that for stack diameters of one to two feet, the plume spreads out to approximately the same size, irrespective of the stack diameter. Also, for a given stack diameter, it is evident that the dilution of the plume at the ground increases as the stack gas velocity and, consequently, as the total contaminant emission rate increase.

It is probable that the indicated dilutions are more than one would expect for descending plumes. Church,[19] for example, obtained least dilutions of 200 in the plume at the ground for a discharge with approximately the same specific gravity as the ambient air. This was for an unstable atmosphere and a 16 inch diameter stack, 200 feet high. This dilution is for about a 10 to 20 second interval, according to Fuquay.[20] The estimated dilutions in Figs. 6 and 7 are one minute averages. This difference in time intervals probably accounts for some variation between the estimated dilutions of this study and the measured dilutions of Church's. In addition, another significant part of the difference is due to the relative motion and the resulting induced mixing of the dense plume and ambient air. On the other hand, for the unstable atmosphere in which Church obtained a least dilution of about 200 for neutral buoyancy, the plume is caught in an eddy and both travel earthward together at the same speed. The dilutions of Figs. 6 and 7 are significantly less than would be obtained, however, if least dilutions at the ground were computed by meteorological dispersion equations, for example, the Bosanquet-Pearson equation.[1] At the location of maximum ground-level concentration, if it is assumed the plume

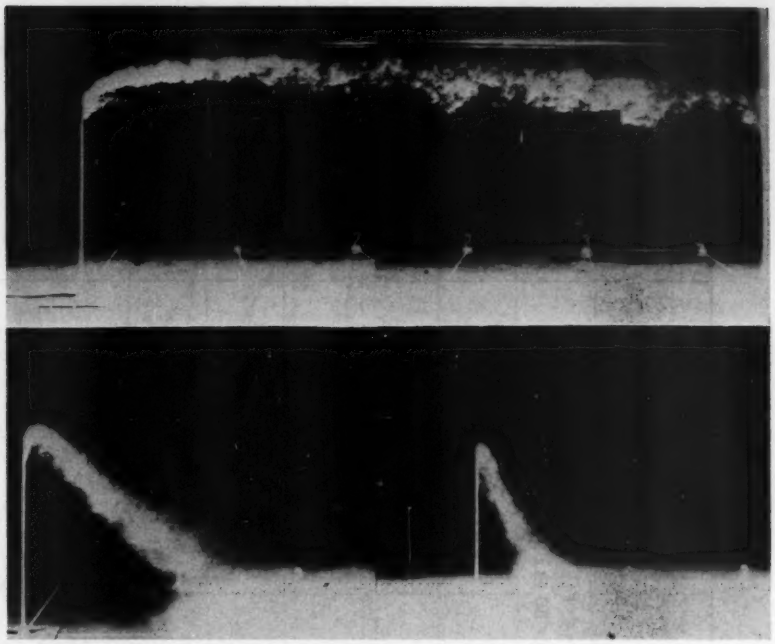

Fig. 1. Photographs of plume behavior. Wind speed, 3 mph; stack gas velocity, 20 ft/sec; stack diameter, 2 ft; stack height, 100 ft. Upper photo, specific gravity = 1 (air). Lower left photo, specific gravity = 1.52. Lower right photo, specific gravity = 5.17.

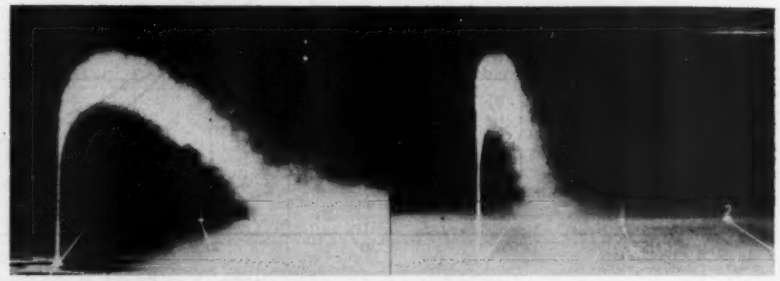

Fig. 2. Photographs of plume behavior. Wind speed, 3 mph; stack gas velocity, 80 ft/sec; stack diameter, 2 ft; stack height, 60 ft. Left photo, specific gravity = 1.52. Right photo, specific gravity = 5.17.

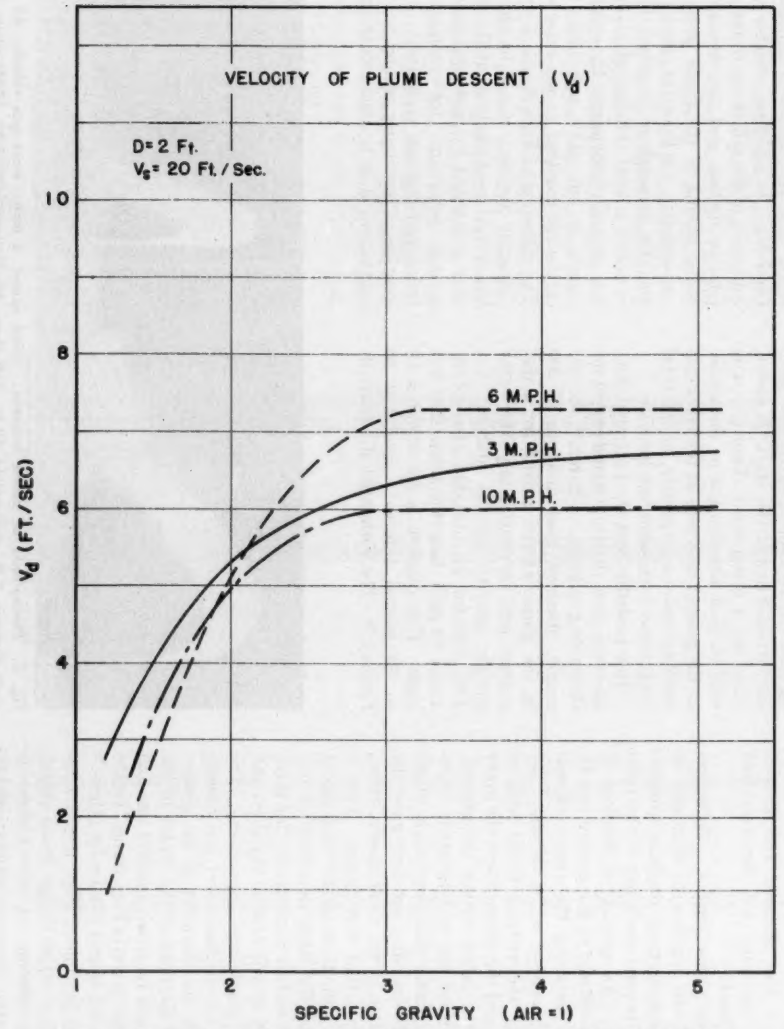


Fig. 3. Velocity of plume descent vs specific gravity for stack gas velocity of 20 ft/sec.

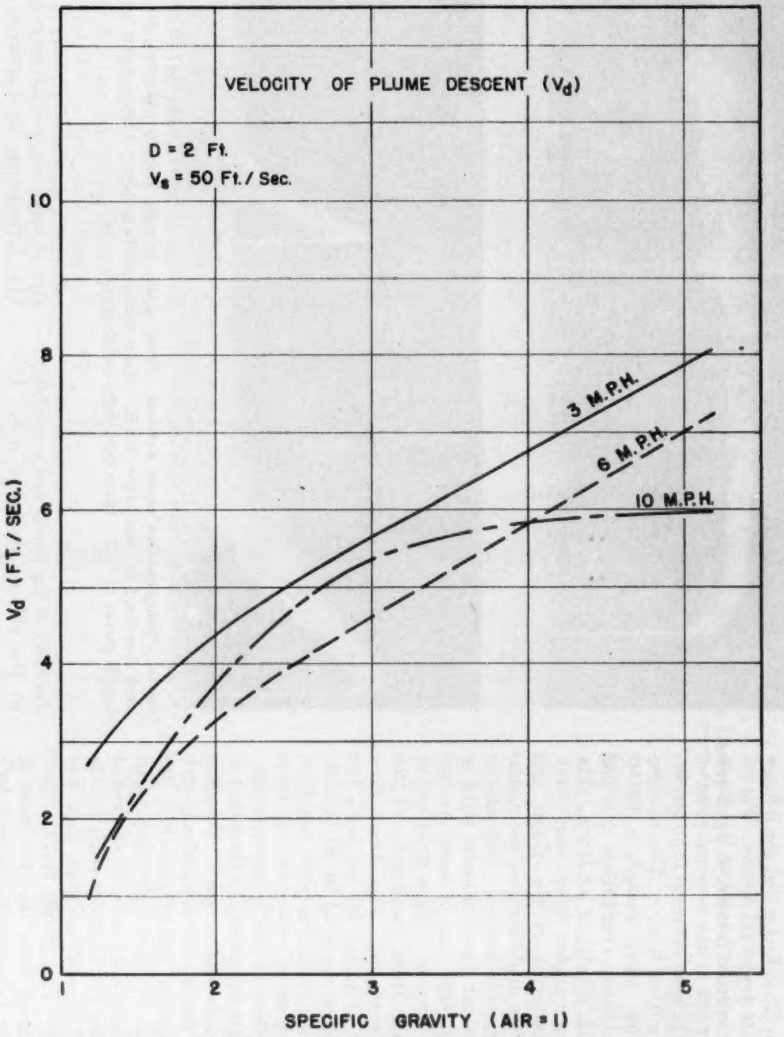


Fig. 4. Velocity of plume descent vs specific gravity for stack gas velocity of 50 ft/sec.

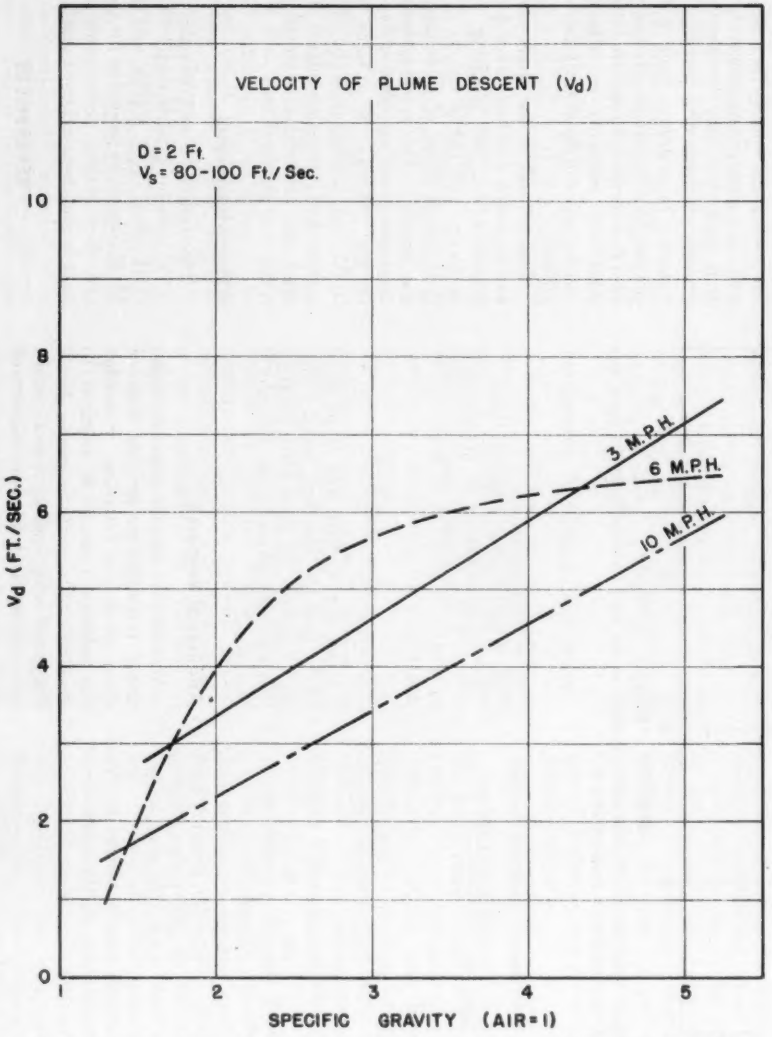


Fig. 5. Velocity of plume descent vs specific gravity for stack gas velocity of 80–100 ft/sec.

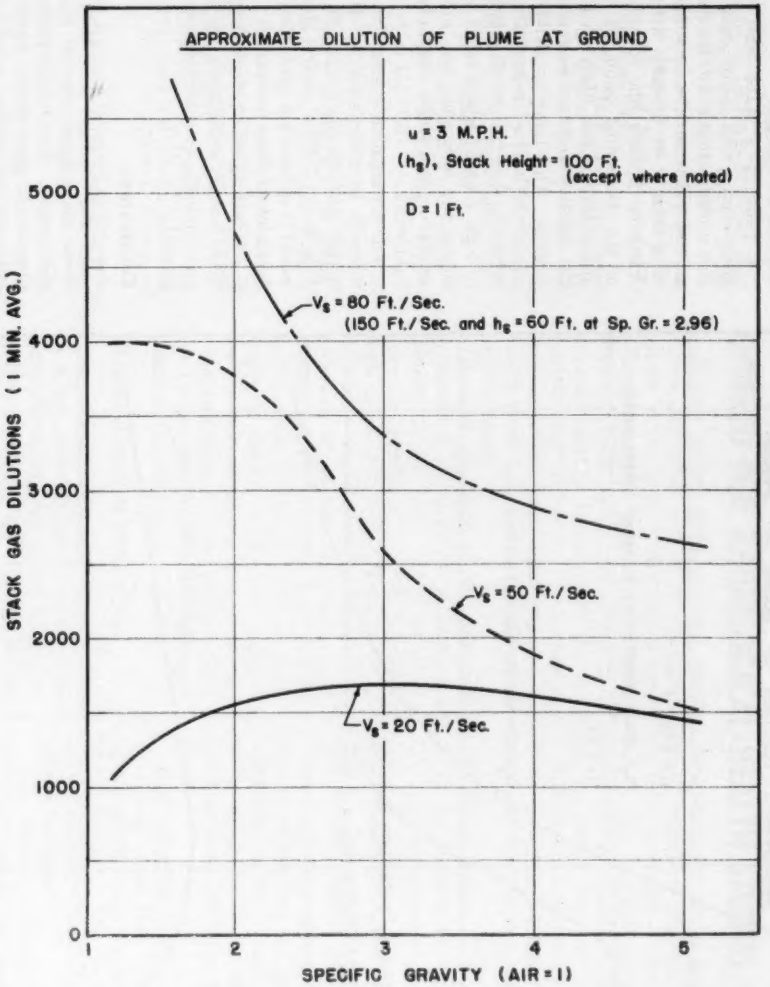


Fig. 6. Approximate dilution of dense stack gas plume at the ground vs specific gravity for a wind speed of 3 mph and a stack diameter of 1 ft.

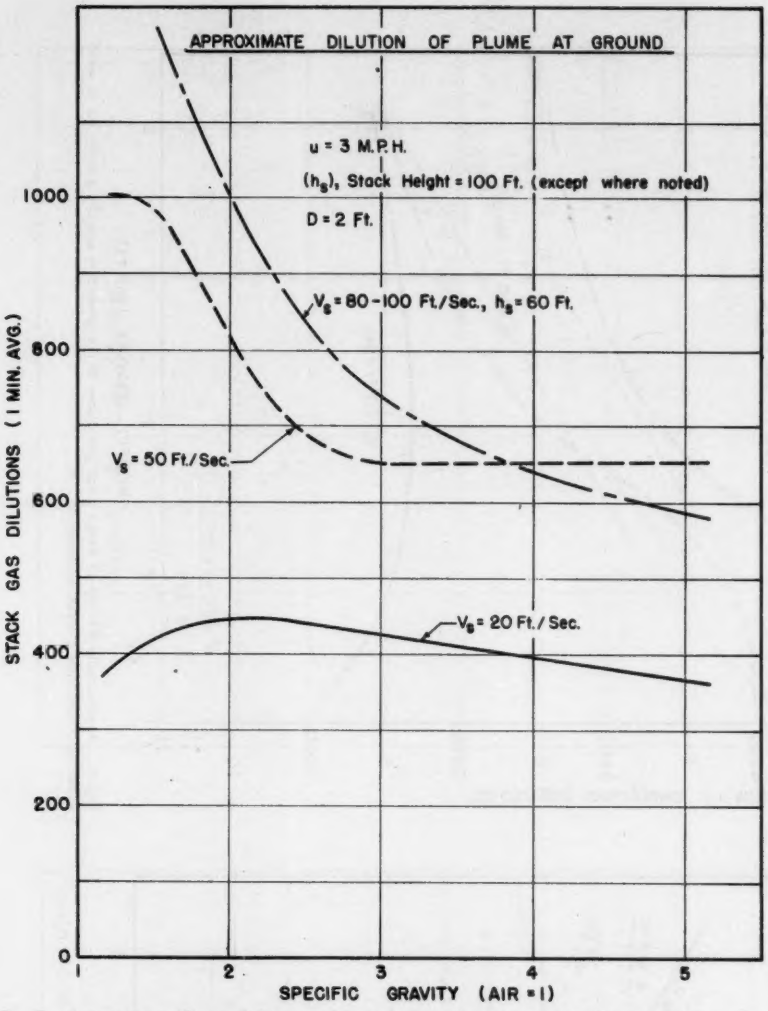


Fig. 7. Approximate dilution of dense stack gas plume at the ground vs specific gravity for a wind speed of 3 mph and a stack diameter of 2 ft.

does not descend,

$$C_0 \text{ Max.} = \frac{2.15\, Q(p/q)\, 10^5}{uH^2} \quad (3)$$

where:

$C_0$ Max. = maximum ground-level concentration (ppm by volume)
$Q$ = contaminant emission rate, cu ft/sec
$p, q$ = dispersion coefficients, dimensionless
$H$ = "effective" height of emission, comprised of stack height, $h_s$, plus rise of plume above stack tip

This formula can be altered to give the least dilutions at the ground, as follows:

$$d = \frac{C_s}{C_0 \text{ Max.}} = \frac{uH^2}{0.215\, Q_s(p/q)} \quad (4)$$

where:

$d$ = least dilutions
$Q_s$ = *total* stack gas flow, cu ft/sec
$C_s$ = concentration of contaminant in the stack, ppm by volume

since:

$$Q = Q_s C_s\, 10^{-6} \quad (5)$$

Using $p/q = 0.63$[7] and $H$ as stack height plus the observed initial plume rise, equation (4) gives least dilutions that are much greater than indicated in Figs. 6 and 7.

## Additional Research

We have no present plans for additional investigation in this field of dense stack gas behavior. Nevertheless additional study is required to describe this behavior in greater detail. Suggested additional data from model tests and/or field tests for some stack and atmospheric parameters are indicated below.

*1* Measurement of concentrations, utilizing a tracer gas.

*2* Tests in a stable and unstable atmosphere with near calm winds. It is probable that concentrations at the ground will be greater in a stable than in a neutral or unstable atmosphere. Adiabatic heating due to compression of the descending plume in a stable atmosphere is not likely to overcome the negative buoyancy from usual stack heights. This, however, could be determined in future tests. The writer estimates that the one minute dilutions of Figs. 6 and 7 would be at least two times the dilutions for comparable conditions in a stable atmosphere with light winds.

*3* Determination of least dilutions for intervals of a few seconds. It is estimated that the one minute dilutions of Figs. 6 and 7 for a neutral atmosphere would be at least five times the dilutions for an interval of a few seconds.

*4* Effect of other stack heights and diameters on dense stack gas behavior and dilutions, or concentrations, at the ground.

## Conclusions

The present study demonstrated that dense stack gases descend rapidly to the ground. The results indicate that dense stack gases should be discharged vertically upward from a stack with the minimum practicable diameter. This minimum diameter results in the maximum stack gas velocity for a given flow rate. In some cases, however, collection equipment may be necessary to prevent pollution or a potential safety hazard. In addition, these tests emphasize that previously-existing atmospheric dispersion equations cannot be used with reliability to estimate the concentration of dense stack gases at the ground.

Sutton[10] and Scorer[12] have discussed the importance of high stack gas temperatures and the resulting positive buoyancy and plume rise in reducing the concentrations of contaminants at the ground. The present study confirms the governing effect of buoyancy on the behavior of a stack plume. Additional investigation of this important and interesting subject of dense stack gas behavior is required to describe it in greater detail.

## Acknowledgments

The assistance and co-operation of NYU personnel, particularly Professor G. H. Strom, E. J. Kaplin, and the late M. Hackman, in preparing for the tests and their skillful performance of the tests is gratefully acknowledged.

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# EXPANDED PROGRAM FOR RESEARCH GRANTS OFFERED FOR RADIOLOGICAL HEALTH STUDIES

In view of the rapid expansion which is forecast in the use of nuclear energy, x-rays, and other sources of radiation, a greatly expanded program for research grants in the field of radiological health has been developed. These grants are offered to support research by individuals, universities, hospitals, laboratories, and other public or private institutions in the assessment and control of man-made and natural radiation exposures to the individual, no matter how the separate components may originate. The knowledge and skills of many professional disciplines and specialties—physicians, engineers, physicists, chemists, educators, statisticians among them—are needed to find answers to the many challenging questions in radiological health.

Research proposals should contribute to the determination of the extent and character of the radiation problem as well as the mechanisms by which radiation damage "cause and effect" relationship are essential if low-level and long-term radiation exposure effects are to be accurately assessed and general control programs organized. Therefore, basic studies relating to critical body organs and systems, preferred metabolic pathways for specific radioactive contaminants, and an understanding of the radiosensitizing and modifying effects of various materials are encouraged through the program.

### Studies Aimed at Evaluation

Broad epidemiological studies aimed at a scientific evaluation of the long term effects such as aging, congenital malformations, genetic effects, behavioral patterns, and cancer induction are also of primary concern. Field studies of the movement of radioactive contaminants in biota and human food chains are of special interest, since we know the physical environment may be greatly altered by biological activity, as for example, the concentration of waterborne radionuclides in microorganisms and fish.

### Equipment, Techniques Mandatory

Purely physical studies, such as chemical mechanisms in radiation chemistry, the design of equipment and the development of techniques to accurately assess or reduce the population dosages are mandatory for a successful research program.

Studies aimed at directing scientific findings toward control devices or procedures are necessary in a "total view" of man's ecological system, as are studies that attempt to assess the relationship between health hazards created and possible benefits derived by radiation usage. The determination of the consequences or radiation exposure for present and future generations will require intensive investigation.

For information and/or application forms please contact: Dr. Paul F. Hahn, Chief, Office of Extramural Grants, Division of Radiological Health, U. S. Public Health Service, Washington 25, D. C.

# DUST CHIEF CAUSE OF SANTIAGO SMOG

Santiago, Chile (Reuters)—Santiago has a smog problem, and one of the main ingredients in the blanket of haze that often hangs over the city is dust sweeping down from the Andes Mountains.

This dust is due to the erosion caused by strong winds that blow at heights above 4250 ft where vegetation is scarce or nonexistent. The foothills of the Andes range are only about 25 miles east of Santiago.

The National Health Service attributes to smog and suspended dust 80% of the respiratory diseases of an allergic type, which are constantly increasing, according to statistics provided by hospitals and private doctors.

According to the health authorities in this capital city of 2,000,000 inhabitants, atmospheric contamination by dust is between five and six times above the level considered permissible in a city. As to the smog itself, the rate of contamination is already above general tolerance.

### Cancer Warning Issued

Doctors have noticed an increase in the death rate because of atmospheric pollution, especially among old people, asthmatic children, and heart cases. The Institute for the Prevention of Cancer has warned citizens that the likelihood of contracting lung cancer in Santiago now is greater than it was 10 years ago.

According to Ricardo Haddad, an engineer in the hygiene and labor medicine section of the National Health Service, the abnormal atmospheric pollution recorded in Santiago has shown that the main causes of the high concentration of smog are industrial and domestic activities and transport.

Santiago's geographical position, surrounded by mountains, without periodic rain to cleanse the atmosphere and without strong winds to carry away the by-products of combustion that con-

(*Continued on p. 440*)

# NEW TYPE OF STEEL NOW DEVELOPED; MAY MAKE ANTI-SMOG DEVICES REALITY

A new type of stainless steel for automobile mufflers looks like it might bring anti-smog devices closer to reality—quicker.

Most major metropolitan areas, and particularly Los Angeles, are plagued with a form of smog caused by the emission of unburned fuel (hydrocarbons) from motor vehicle exhausts.

California already has statutes on the books for the use of new devices which will consume practically all of this unburned fuel. While these laws are on the books at the present time, they do not become effective until at least two devices have been approved by the Motor Vehicle Pollution Control Board. This new muffler-like item is called an anti-smog device.

While other states besides California are talking about such devices for cars, they are taking a "wait and let's see what happens in California" attitude.

The new stainless steel, called MF-1, was introduced recently by Allegheny Ludlum Steel Corporation and was developed for use in automotive mufflers, a close, but not as hard-working, cousin of the anti-smog device. Both mufflers and anti-smog devices have much the same problems—corrosion and heat. MF-1 combats both of these problems and is a relatively inexpensive metal.

Because of space limitations, an anti-smog device will likely replace the conventional acoustic muffler. The resistance to corrosion and heat of MF-1 and its attractive price makes this material an ideal choice for anti-smog devices as well as for mufflers.

The California State Board of Health has established limitations of 275 ppm of hydrocarbons, and 1½% by volume of carbon monoxide in the exhaust of automobiles. This requires an anti-smog device to do a big job for a long period of time.

### Removal Methods Described

There are two methods of removing hydrocarbons and carbon monoxide from the exhaust gases of automobiles. One is the catalytic type which passes the exhaust gases through a bed of chemically-treated pellets before emitting them to the atmosphere. This system has low initial cost, but suffers a gradual deterioration in activity, and requires periodic replacement of the catalytic pellets.

The other type is the direct-flame afterburner which employs high temperature to burn the hydrocarbons and carbon monoxide contained in the exhaust gases. This system has a higher initial cost, but is not subject to performance deterioration and, therefore, does not need periodic replacement of any components.

The catalytic system would operate between 800° and 1500°F, whereas the temperature of the flame-type afterburner would be in excess of 1500°F.

Allegheny Ludlum's MF-1 stainless steel is being tested for use in both catalytic and direct flame afterburner anti-smog devices. In some of the tests the stainless steel parts make up virtually the entire device; in others only the most corrosive parts are made of stainless steel.

If all cars were properly adjusted and maintained, and if these cars were operated at steady cruise, there would be no need for anti-smog devices. But, most automobiles operate fuel-rich over a wide range of driving conditions, including idle, acceleration, and deceleration as well as cruise. When this happens, hydrocarbons and carbon monoxide are spewed from automobiles into the air which we breathe.

One firm, which requests that it not be named at this time, has developed several models of motor vehicle pollution control devices. Engineers for this firm have been able to control the exhaust contaminants of motor vehicles equipped with such devices within the limits prescribed by the State of California. Prototypes of catalytic anti-smog devices made of MF-1 steel are now undergoing extensive road testing. The catalytic pellets within the device would be replaced annually or once every two years. Other components of the device would last the life of the car.

## *Giant Collector Ordered*

A giant Therm-O-Flex high temperature glass bag filter type dust collector has been ordered by the Riverside Cement Company, division of American Cement, for their Oro Grande California plant.

The filter will clean the kiln gases and will utilize over 240,000 sq ft of glass cloth. There will be five buildings housing the bags and all will feature precast concrete shells, internal partitions, and supports.

The Therm-O-Flex will be built by Western Precipitation Division of Joy Manufacturing Company, specialists in dust control equipment. The Oro Grande Therm-O-Flex is the largest ever planned and is the ninth such installation made by Western Precipitation in the cement industry—three times as many as all other makes combined.

# Personalia

**Gerard E. Smith** has been promoted to Vice President in Charge of Sales, Western Hemisphere, and has been elected to the Board of Directors of Airkem, Inc., at the Company's Board Meeting in its New York Headquarters, 241 E. 44th St., New York City, as announced by Frank W. Conkling, President of Airkem, Inc.

Mr. Conkling also announed the appointment of **Frederick T. Warner** as Assistant Vice President, Administration, and **Claude J. D'Angio** as Assistant Vice President, Research.

The three officers have been named to membership on the President's Advisory Committee. These moves are designed to strengthen the Airkem Management Team by the addition of Sales-and-Research-oriented.

* * *

**Arvil B. Pettit,** formerly Director of Industrial Health and Safety for W. R. Grace and Company has joined William R. Bradley and Associates, environmental health consultants, as a consultant on safety, fire and explosion, and marine hazards.

While at W. R. Grace and Company, Mr. Pettit had complete responsibility for industrial health and safety, including fire prevention and air and water pollution control of more than 70 industrial plants and mines in the United States and abroad.

During the period in which he worked for Grace's Davison Chemical Division that company received the Award of Honor of the National Safety Council five times in a six year period. In addition, Mr. Pettit has participated in many state and local conferences on industrial health.

* * *

**Charles N. Lockwood** has joined the Alamask Division of Rhodia Inc., New York, leading manufacturer of odor control chemicals and allied products. Mr. Lockwood, former sales manager of Cloroben Chemical Corporation, has had broad experience in the fields of air pollution, sewage, and industrial waste. His major responsibility will be to assist Rhodia's Sales Department in broadening its distribution system through agents.

The appointment of **Schaeffer E. Specht** as General Sales Manager for the Buell Engineering Company, New York, New York, was recently announced by J. L. Schumann, President.

In his new capacity, Mr. Specht will supervise all sales operations for Buell Engineering Company and its Northern Blower Division in Cleveland, Ohio. He was formerly Western District Sales Manager.

## MVP CONTROL BOARD HOLDS 9TH MEETING; REPORTS PRESENTED

The following is a summary of actions taken by the Motor Vehicle Pollution Control Board at its ninth meeting held May 19, in Oakland, California.

*1* These reports were presented:

Organization and Program of the Bay Area Air Pollution Control District. Honorable Francis Dunn, Jr., Member of Board of Supervisors, Alameda County; Member of Board of Directors, Bay Area Air Pollution Control District.

Effect of Motor Vehicle Traffic Upon Production of Urban Air Pollution. Film, courtesy of Los Angeles County Air Pollution Control District.

Report on Air Monitoring in California. George Taylor, Supervising Engineer, Bureau of Air Sanitation, State Department of Public Health.

*2* Authorized the Chairman to draft an appropriate resolution indicating Board support for a statewide air monitoring program as necessary for effectuation and evaluation of the Board program.

*3* Conducted a public hearing on proposed regulations relating to certification of crankcase emission control devices for inclusion in the Administrative Code. After editorial revisions directed the Executive Officer to submit a revised draft of the Order Adopting Regulations of the Motor Vehicle Pollution Control Board Pertaining to Approval of Motor Vehicle Pollution Control Devices which describe requirements for certification of crankcase emission control devices. The Board members were to approve the regulations by mail vote on May 24, 1961.

*4* Re-elected Dr. John T. Middleton and James Allen as Chairman and Vice Chairman of the Board for the fiscal year starting July 1, 1961.

*5* Heard the report of the Committee on Test Procedures and Their Evaluation relative to the second draft of the test procedures for exhaust control devices. After discussion by the Board members and comment from the audience, the test procedures were approved as submitted.

*6* Hard report from Budget Committee on a draft of a contract with the Los Angeles County Air Pollution Control District Automotive Laboratory for the fiscal year commencing July 1, 1961. Authorized the Executive Officer to sign such a contract after it was in final form following clearance with the Budget Committee and the Executive Committee.

*7* Accepted a report on a public information program submitted by the Executive Officer.

(*Continued on p. 443*)

## *RESEARCH-COTTRELL PUBLISHES BROCHURE ON CONTROL EQUIPMENT*

The latest techniques and equipment for air pollution control are presented in a new 12-page illustrated brochure published by Research-Cottrell, Inc., leading U. S. manufacturer of industrial gas cleaning equipment.

New air-cleaning precipitators for nuclear subs, automated controls, and high-efficiency silicon rectifiers are shown as recent products of the modern research, design, and manufacturing facilities available at Research-Cottrell.

Recent installations of high-performance gas cleaning equipment in key industries such as basic oxygen steel plants, paper mills, electric utilities, cement and chemical plants are also given from among the 2400 Cottrell precipitators sold, the largest number for any company.

Research, laboratory, testing, and service facilities available to customers are also listed and described.

Copies of Bulletin 500 "This Is Research-Cottrell" may be obtained by writing to Research-Cottrell, Inc., Bound Brook, N. J.

## *In Memoriam*

We regret to announce the loss of two of APCA's members: Desmond P. Tynan, died June 14, and F. Forrest Pease, died June 25.

## LETTERS TO THE EDITOR

(*Continued from p. 402*)

Editor:

I have noted with interest the article on the John Wood Company booklet "Together We Can Check the Blight of Air Pollution," which appears in the June 1961 issue of the Journal of the Air Pollution Control Association.

I do not question the right of the John Wood Company to publish and distribute this booklet. However, I do question the appropriateness of the APCA article about the booklet (June 1961 JOURNAL) with the implied endorsement of its accuracy and of the positions taken on a number of unsettled problems and debatable issues.

The official opinion of the U. S. Department of Health, Education, and Welfare of the Public Health Service is contained in a letter from Assistant Secretary Wilbur J. Cohen to Mr. Loree Collins of the John Wood Company, dated May 5, 1961. A copy of Mr. Cohen's letter and of Mr. Collins' reply are enclosed. It is suggested that these two letters be published together in the Journal.

V. G. MACKENZIE, *Chief*
Division of Air Pollution

## BAY AREA BOARD ADOPTS AMENDMENTS

Amendments to the Administrative Code which create a new position of Chief Administrative Officer were adopted by a unanimous vote of the Board of Directors of the six-county Bay Area Air Pollution Control District, at a recent meeting.

Nine members of the 12-member Board were present to vote for the changes, which are anticipated to go into effect by October 1, 1961. The new Chief Administrative Officer will be appointed by the Board and be responsible to them for the general operation of the District staff, while Air Pollution Control Officer Benjamin Linsky will remain responsible for technical and enforcement division activities.

New officers for the coming fiscal year 1961–62 were also elected as follows: Chairman, Supervisor Clarissa Shortall McMahon, San Francisco; Vice Chairman, Clarence D. Erickson, City Council of Richmond; re-elected Secretary, Warren G. Buchanan, Supervisor, Contra Costa County. Outgoing Chairman was Andrew C. Byrd, Mayor of Burlingame, and Mrs. McMahon had been Vice Chairman.

In other business, Control Officer Benjamin Linsky announced that the U. S. Public Health Service has offered to provide an engineer to the District as an air pollution control trainee at no charge. The Board approved the suggestion.

Committee reports were made regarding budget and finance, technical services, legislation, hearing board cases, and enforcement procedures.

### Welcome New Members

COMPANY MEMBERSHIPS (NATIONAL)

Armstrong Cork Company
Lancaster, Pennsylvania

INDIVIDUAL MEMBERSHIPS

Baldwin, John E., Hollywood, California
Berman Leonard, Cleveland, Ohio
Bondra, Stephen, Cleveland, Ohio
Corn, Morton, Pittsburgh, Pennsylvania
Gjerde, R. K., Tacoma, Washington
Hersey, Mrs. Henry J., Jr., Chatham, New Jersey
Johnson, John E., Buffalo, New York
Lake, Herman, Dedham, Massachusetts
Lewis, Frederick B., Everett, Massachusetts
Peck, Dr. R. DeWitt, Montgomery, West Virginia
Selvidge, Dr. Harner, Altadena, California
Thomas, W. C., Nichols, Florida
York, Loftus S., East Pittsburgh, Pennsylvania
Zachow, Arnold A., Philadelphia, Pennsylvania

# NEWS FROM THE CONTROL DISTRICTS

### New York City

Criteria for the installation of new flue-fed incinerators in New York City, which should result in improved design and less air pollution from this source, have been put into effect by the New York City Department of Air Pollution Control, it was announced by Commissioner Arthur J. Benline. The criteria, which represent several years of work by the Department in co-operation with engineers, architects, manufacturers, and control officials, took effect July 1.

Flue-fed incinerators are the type used in most apartment house buildings in New York City. It is estimated that there are more than 11,000 in use, with about 8000 having been in existence since before the Department was established in November 1952.

While the rules and regulations of the department have caused improved design during the past eight and a half years, the necessity was found for interpretations of the rules as far as dimensions and capacities are concerned. The criteria now in effect are designed to serve that purpose.

Among the features of the new criteria are requirements that where the flue is used as a chute as well—the situation of most apartment house incinerators—the hopper doors are to be provided with locks which operate during the burning cycle. This will prevent the depositing of refuse at a time when it would interfere with the combustion in the incinerator chamber. It is also a safety measure, since it prevents a tenant being burned accidentally during the burning cycle.

Another feature requires that incinerators in buildings with more than 500 rooms be completely automatic in operation. This requirement is necessary because manual operation of incinerators in very large buildings has been unsatisfactory.

Use of double flue incinerators—with the chute separate from the combustion flue—is encouraged by the criteria, since this type of incinerator results in less air pollution than the single flue-fed incinerator.

### Benline Comments

Commenting on the new criteria, Commissioner Benline said: "These criteria represent the work of many people, over several years. We have been pleased to receive suggestions and criticisms of our proposals. We believe that although perfect incineration will not result from these criteria, this is an important development toward making this device much more acceptable from an air pollution control standpoint. As experience and technological progress dictate, the criteria will be improved and refined, to take advantage of the best possible designs."

Operating and maintenance instructions, as well as several charts, tables, and drawings are included as part of the criteria, for the guidance of those designing, manufacturing, constructing, and operating incinerators. Copies of the criteria may be obtained from the Bureau of Engineering, Department of Air Pollution Control, 15 Park Row, New York 38, N.Y.

## SANTIAGO AP PROBLEM

(*Continued from p. 437*)

taminate the air, or the dust from the Andes, have placed the city in a position that is becoming increasingly critical.

The first steps to remedy the situation are being concentrated on the worst areas of contamination.

### Diesel to Be Barred

All industries are being listed, and their owners are being required to install special filters. With the help of the police and municipal authorities, private and public vehicles are to be compelled to keep their engines in good order, so as to reduce the quantity of exhaust gases. All vehicles using diesel oil as all or part of their fuel will be withdrawn from circulation.

Simultaneously with a decree forbidding the establishment of new industries within the city limits and new health regulations for the installation of industries outside the city, the municipality also is studying various projects to give industrial uses to garbage.

Until now, tons of garbage from the city's dust bins are carried to vacant spaces outside Santiago and burned there in the open air. This, too, in certain circumstances and with the wind's always willing assistance, subjects some of the quarters of Santiago to nauseating smells during the night when the disposal of garbage takes place.

A decree issued by the Ministry of Public Health applies not only to Santiago, but also to the provinces, and provides penalties for offenders.

It forbids the open incineration of garbage within the city limits whether in the street or on private property as well as the circulation of every vehicle with a visible column of exhaust gas. Offenders will be liable to penalties under the health code.

Another article in the decree sets deadlines for the installation of filters or other purifying devices.

# NOMINATIONS NOW OPEN FOR APCA'S 1962 AWARDS

Nominations are in order for people to receive honors or awards given out each year by the Air Pollution Control Association. These nominations constitute one of the most important activities of the year, since they promote professional interest in the accomplishments of fellow members and encouragement to all to contribute to the betterment of the objectives of the Association.

First award to be established by the Association was the Frank A. Chambers citation "for outstanding achievement in the control of air pollution" and dates back to February 12, 1954. Eligibility for this award goes beyond general excellence of work and prominence in the field into the tangible achievement in design of equipment, investigation, and determination of phenomenon not previously understood, or in a scientific application of existing equipment to solve situations we never before attempted.

The second award, named in honor of Richard Beatty Mellon, was established in 1955 to honor an individual "whose contributions of a civic, administrative, technical or other nature have aided substantially the cause of air pollution control." This award can be made to members and nonmembers of the Association. Consisting of a certificate, this award cites the service given by an individual over a period of time to develop the acceptance of the cause of air pollution.

### Honorary Membership

All members are encouraged also to recommend as candidates for honorary membership names of people of eminence in the field of air pollution control or who have rendered especially meritorious service to the Association. Those accorded this recommendation will become honorary members of the Association, having the privilege of attending meetings and other privileges of the floor, except they do not have voting privileges.

All nominations and recommendations must be accompanied by a full and detailed account of the accomplishments upon which the nomination is based. It should also be accompanied by appropriate personal and background material concerning the candidate. Five references with letters of support are required.

Nominations should be sent to Dr. Louis C. McCabe, Chairman of the Awards Committee, or to any of its members not later than January 1, 1962. Committee members are: Dale H. Hutchison, Charles W. Gruber, Herbert C. McKee, and Dr. L. V. Cralley.

## PAUL CROSS APPOINTED MELLON INSTITUTE HEAD

Dr. Paul Clifford Cross of the University of Washington, Seattle, has been named President and Chief Executive Officer of Mellon Institute. He also was elected a Trustee of the Institute.

Paul Mellon, Chairman of the Institute's Board of Trustees, made the announcement following a meeting of the Board.

Dr. Cross, a native of Bruin, Butler County, Pennsylvania, has been Professor and Executive Officer of the Department of Chemistry of the University of Washington since 1949. He assumed his new responsibilities at Mellon Institute, September 1.

Dr. Cross was born July 19, 1907. He received his bachelor's degree from Geneva College in 1928 and his doctor's degree from the University of Wisconsin in 1932. He subsequently held research fellowships at the latter university, California Institute of Technology, and Stanford University. He became an assistant professor at Stanford in 1936, and in 1938 joined Brown University as an associate professor. From 1942 to 1949 he served successively as Professor Director of the Metcalf Research Laboratory, and Chairman of the Chemistry Department at Brown.

### Long Research Career

During his career in research and education, covering nearly 30 years, Dr. Cross has been associated with numerous military scientific activities. From 1940 to 1943 he was a consultant on various war projects at Brown University. In the period 1943–44 he was Deputy Director, Underwater Explosives Research Laboratory, OSRD, at the Woods Hole Oceanographic Institute. He was Research Director of the latter from 1944 to 1946.

Dr. Cross has also been a summer lecturer at Harvard and the University of Wisconsin. He is a member of numerous professional and honor societies and is a contributor to the journals. In 1932 Dr. Cross married Sara A. Groves, of Kaylor, Armstrong County, Pennsylvania. They are the parents of two sons and two daughters.

**Help Boost APCA Membership**

## CLEANER AIR WEEK SCHEDULED OCTOBER 22–28; KEYNOTES INDIVIDUAL RESPONSIBILITY IN APC

Cleaner Air Week, sponsored by the Air Pollution Control Association, is being held this year from October 22 to 28.

CAW is the brain child of Charles Howison, executive secretary of Greater Cincinnati's United-Fund supported Air Pollution Control League, and air pollution control pioneer.

### The Objective

Cleaner Air Week's threefold objective is as follows: public understanding of the community's air pollution problems, progress being made toward their solution, and the importance of the community-wide control approach.

Everybody gets in the act during Cleaner Air Week with all interests in pollution problems represented on the local CAW Committee and participating in every phase of its activities.

The local control official frequently heads the local group, organized with the help of procedures provided by Charlie Howison's National Cleaner Air Week Committee.

This volunteer group makes maximum use of news public service, advertising and public speaking opportunities to "sell" the individual citizen his stake in the community's cleaner air program and the need for continued co-operative effort to protect his most precious asset —clean air.

### The Program

Mr. Howison's National Cleaner Air Week Committee, beginning in June, issues periodic bulletins to local cleaner air week committees and interested groups, covering everything from committee organization to staging plant tours for inspection of new air pollution control equipment.

Suggested speeches, news releases, radio and TV interviews; with emphasis on "Project Air Lift," perennial balloon ascension stunt, make up the substance of the program. College football games star "Project Air Lift" during their half time programs.

### The Keynote

The ultimate "order" which Cleaner Air Week seeks is the individual's acceptance of his role in cleaning up our air and his realization that effective control progress can be made at all segments of community life working in co-operation.

Most Cleaner Air Week costs are buried in the public relations department of government agencies, academic groups, and industries participating in the program. It succeeds on pennies because of its universal appeal. Whatever he does for Cleaner Air Week, John Doe knows he's doing those things for himself.

Eighty metropolitan areas are now enlisted in Cleaner Air Week ranks and the rate of growth continues to rise. As more and more communities participate, the trend is to individual programs, which are tailored to local control problems.

## *Four Men Appointed For Trans-atlantic Call*

Dr. Weidlein Dr. Prindle

Dr. Faith Mr. Arch

Four men have been appointed to take part in the Trans-atlantic telephone call to be held during the 28th Annual Conference of London's National Society for Clean Air. They are: Dr. Edward R. Weidlein, scientist, author and retired president of Mellon Institute; Dr. Richard A. Prindle, Medical Director and Chief of Air Pollution Medical Program; Dr. W. L. Faith, Chemical Engineering Consultant and President of APCA; and Mr. Arnold Arch, Executive Secretary of APCA.

The call is scheduled to take place October 5 either in Pittsburgh or New York.

Dr. Weidlein will act as chairman of the four-man team which will hold an unscripted question and answer discussion with members of the society in London for 30 or 40 minutes. This feature is the hi-lite of the meeting and is sponsored by the Pfizer Company, Inc.

# VEHICULAR EXHAUST COMMITTEE MINUTES

The Vehicular Exhaust Committee of APCA met during the Annual Meeting on June 12 at the Hotel Commodore, New York City. The minutes of that meeting are published below.

Attendance:
Dr. W. L. Faith, President, APCA
Harry Belyea, Past-President, APCA
S. Smith Griswold, Chairman
Mrs. Stanley Feuer
Leo P. Flood
Charles Howison
Roger W. Sackett
Arthur A. Atkisson, Secretary
Charles M. Heinen (Absent)
Andrew H. Rose (Absent)

The committee meeting was opened by Mr. Griswold who introduced committee members and outlined the importance of the committee's role in the APCA program. Suggestions received from committee members for future committee activities were discussed in detail, with reports from Messrs. Flood and Howison.

The range of discussion included the following:

*1* Need for preparation of a report for the general information and guidance of APCA members, the report to deal with such specific matters as:

- *a* Relationship between vehicular emissions and deterioration of air quality.
- *b* Circumstances under which alternative levels of vehicular contaminant control efficiencies may be required to improve or protect community air quality.
- *c* The general scope of the vehicular emission problem in the nation today and in the forseeable future.
- *d* Present research and control-development activities being pursued by public and private agencies.
- *e* Types of controls available or anticipated, and their estimated cost and efficiency. Discussion to include consideration of blowby, deceleration, catalytic, and afterburning devices as well as such other control measures as fuel modification and improved maintenance.
- *f* Safety Lane Inspections, as performed twice yearly by Cincinnati and Norwood, Ohio, for the purpose of detecting and requiring correction of mechanical and functional defects.

*2* The need for a simple means of assessing community air quality trends to determine the existence of a vehicular contamination problem in any given community, or a standard evaluation form to be used by local officials in appraising the existence of community air pollution problems related to vehicular contaminant emissions. It was indicated that such a simple appraisal might not provide definitive answers as to the specific need for control or the efficiency required of such controls, but might well indicate the need for further definitive study in specific communities and the existence of gross control requirements.

*3* The control possibilities suggested by the use of alternative fuels. Mr. Flood raised the specific question of LPG as a substitute for gasoline and/or diesel fuels, at least for selected portions of a community's vehicles. He stated that this particular area of inquiry might well be probed in further detail by the United States Public Health Service.

*4* The possible need for modifications in engine design. Mrs. Feuer raised the question of the possible need for modifications in engine design as a means of curbing vehicular contaminant losses. Further discussion between Mrs. Feuer, Mr. Flood, and Mr. Griswold indicated the value of such a step as a basic and long term approach to the problem. Although more immediate control solutions must be developed, engine redesign to attain greater combustion efficiency remains a desirable and necessary goal.

President Harry Belyea commented on the important role of the Committee in the total APCA program and underscored the view that control officials throughout North America are in extreme need of digested and summarized information regarding vehicular pollution. He suggested that the future committee program should consider the following:

*1* Determination of the needs of APCA members and the ways in which the committee may satisfy those needs.

*2* Performance of a literature review to identify information sources of relevance to the vehicular problem, and the distribution of such a bibliography to the APCA membership.

*3* Identification of specific problems in the vehicular field which require the attention of the Association, and the preparation of recommendations to solve the problems.

### Committee Action

Following discussion of these matters, the committee determined that the following policies and programs should guide future committee activities:

*1* The committee should meet two to four times annually and should file progress and information reports with the *APCA Journal*, following each meeting, as a means of informing the total membership on problems and progress in this field.

*2* A "status report" or "information packet" on the vehicular problem should be prepared for the guidance and advice of the committee and for distribution to the APCA membership. Mr. Griswold agreed that the Los Angeles County Air Pollution Control District would undertake the immediate preparation of such a report, and that it would include a bibliography of information

(*Continued on p. 443*)

# S-4 LEGAL COMMITTEE ANNUAL REPORT

The Legal Committee reports with regret the death during the course of the year of two of its valued and faithful members: Prof. W. W. Hodge of Pittsburgh and Walter S. Cooper of New York. Their able counsel will be missed by the Committee.

We can report the following accomplishments:

(*1*) The Committee has agreed upon and submitted to the Board of Directors for approval and declaration as a policy of the Air Pollution Control Association of a Resolution on procedures relating to the rule making process;

(*2*) An analysis of all reported legal decisions in the air pollution field for the period, December 1959 to May 1961, has been completed and will be set forth in coming issues of the JOURNAL;

(*3*) The Committee submitted a series of by-law amendments relating to the election of officers and directors which were approved by the membership and continues to work with the By-Law Committee in a continuing review of existing by-laws.

The Committee views with some concern the expression of legal opinions by nonlawyers in the air pollution field. With the consent of the technical director of the 1962 meeting, the Committee proposes to put on a legal seminar for control officials as a first step in the correction of this trend.

ERWIN E. SCHULZE, *Chairman*

# CALIFORNIA MAKES STEADY PROGRESS TOWARD APC OF MOTOR VEHICLES

California's steady progress toward control of air pollution stemming from its 7.5 million motor vehicles is reported by the Motor Vehicle Pollution Control Board.

The Board, meeting in its 10th session at the State Office Building, declared that standards for air pollution have now been determined and that manufacturers are at work on devices to control both exhaust and crankcase emissions.

To date, the Board has received 22 applications for control devices. Fifteen of these were rejected as not meeting the requirements of the Board. Of the remaining seven, six applications are now being screened, while one, that of Oxy-Catalyst, Inc., of Berwyn, Pennsylvania, was accepted for Step I testing three weeks ago.

Certification also is pending on one crankcase device, that of the AC Spark Plug Company. The Board reported that automobile makers already are installing this device voluntarily on all 1961 American cars sold in California.

Companies whose applications are now being screened for exhaust control devices are the Universal Oxidation Processes, Inc., Los Angeles; Arvin Industries, Columbus, Indiana; Wolverine Tube Division of Calumet and Hecla, Inc., Allen Park, Michigan; Standard Products Company, Cleveland, Ohio; Chromalloy Corporation, Hawthorne; and W. R. Grace and Company, Baltimore, Maryland.

The Board said that reports of an "elderly breakthrough" in control of exhaust emissions are "encouraging" and "competition in crankcase emission control apparently is forthcoming."

"We do not intend to rush into a hasty decision on these devices," said Dr. John T. Middleton of Riverside, Board chairman, "because of the vast technical field to be covered and the amount of money which California's motorists will have to spend." This sum is estimated in excess of $500 million.

Dr. Middleton said the Board is primarily interested in devices that will be easy to install, perform effectively, and have a long life.

He pointed out that the compliance portion of the law would take effect only after at least two devices for both crankcase and exhaust control have been certified.

Following that would be a three-year schedule for installation of the devices—first on all new cars, then commercial vehicles and used cars when their registration is transferred, and, finally, on all motor vehicles in the state.

Counties that feel they have no air pollution problems as yet will be exempt from the program, provided their situation is reviewed every two years.

The 13-man Board, appointed by Governor Brown, is made up of leaders from industry, labor, science, public health, and law enforcement.

# ANALYSIS OF SMOKE

(*Continued from page 416*)


6. R. E. Munn and M. Katz, "Daily and Seasonal Pollution Cycles in the Detroit-Windsor Area," *Int. J. Air Poll.*, **2**: 51–76 (1959).
7. F. Gifford, "Peak to Average Concentration Ratios According to a Fluctuating Plume Dispersion Model," *Int. J. Air Poll.*, **3**: 253–260 (1960).
8. I. A. Singer, "The Relationship Between Peak and Mean Concentrations," 53rd Air Pollution Control Association Meeting, May 1960.



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# CALENDAR

| | |
|---|---|
| October 4–6 | 28th Annual Conference of National Society for Clean Air, Brighton, England. |
| October 16–17 | International Conference on Ionization of the Air, The Franklin Institute, Philadelphia, Pennsylvania. |
| November 27–28 | Fourth Conference on Agricultural Meteorology, American Meteorological Society in conjunction with American Society of Agronomy, St. Louis, Missouri. |
| February 19–21, 1962 | Clean Air Conference, sponsored jointly by New South Wales Department of Public Health and University of New South Wales, University of New South Wales, Sydney, South Wales. |
| May 20–24, 1962 | 55th Annual Meeting of APCA, Chicago-Sheraton Hotel, Chicago, Illinois. |

# *COMMITTEE MINUTES FROM ANNUAL MEETING*

(*Continued from p. 442*)

sources that might be of interest and assistance to APCA members.

*3* A questionnaire survey will be conducted of all Air Pollution Control Officials to determine their assessment of vehicular contaminant problems, and their needs for information, control programs, etc. Mr. Griswold agreed to prepare a suggested questionnaire for early review by the committee members.

*4* The committee concluded that present evidence suggests the need for nation-wide utilization of "blowby" controls on all new vehicles. Following review of questionnaire responses by Control Officials, and such other evidence as may be relevant, the committee will query the AMA regarding installation of "blowby controls" on all new vehicles.

*5* Upon suggestion by Mrs. Feuer, the committee unanimously directed the chairman to solicit a full statement from the AMA regarding their activities and the present status of control devices.

# SUMMARY OF ACTIONS OF MVP CONTROL BOARD

(*Continued from p. 439*)

*8* Accepted a progress report from the Executive Officer on staff procurement, office facilities, and staff activities, and indicating that the Board has nine pending applications for exhaust control devices and two from crankcase emission control device manufacturers.

*9* Heard a report from the Executive Officer on a conference with the U. S. Public Health Service on pertinent research projects which would be cooperatively undertaken in California in the next fiscal year including an announcement that the U. S. Public Health Service plans to assign a senior engineer to the engineering staff of the Motor Vehicle Pollution Control Board to render technical assistance and liaison functions.

*10* Heard a report on the appointment of a technical advisory group to consider the program on enforcement and compliance.

*11* Heard the announcement by the Chairman that Harold Thompson, a Board member, has been appointed to the Superior Court of Fresno County by Governor Brown and will therefore no longer be able to serve as a member of the Board.

*12* Set the time and the place of the next meeting as 10 AM, July 14, 1961, State Office Building, Room 1122, 107 S. Broadway, Los Angeles.

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# of the

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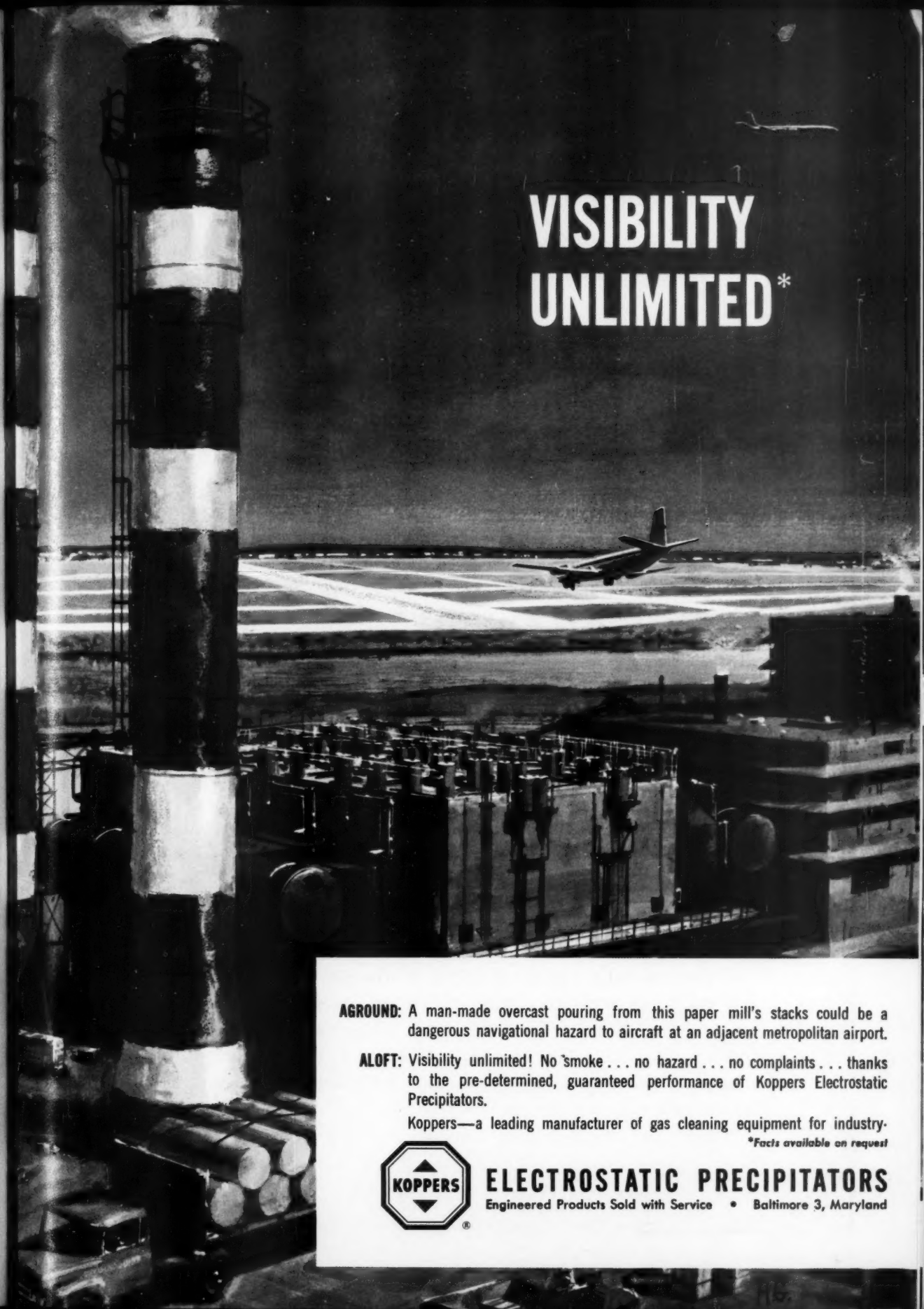
Papers submitted for this meeting may not be offered for publication elsewhere than the *Journal of the Air Pollution Control Association* unless specific written release is obtained from the Association. The Association will, however, give the author not later than three months after the date of presentation of the paper either a firm commitment that the paper will be published in the *Journal* of the Association or a release for publication elsewhere.

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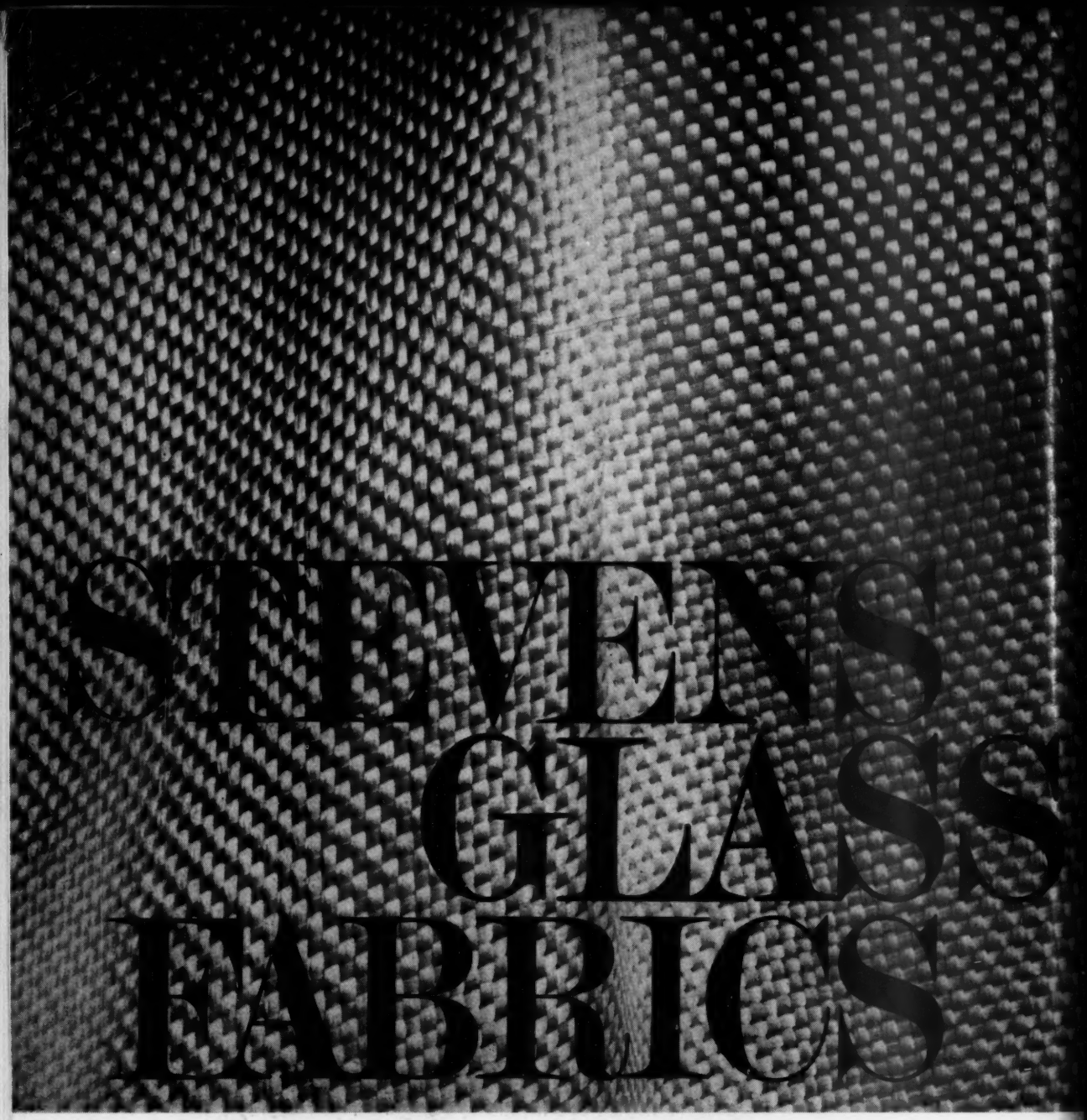


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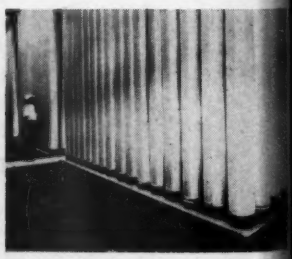

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